OCEAN DISPOSAL OF REACTOR WASTES

Woods Hole Meeting August 1954

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NYO Waste Disposal



Report of Meeting on
OCEAN DISPOSAL OF REACTOR WASTES
Held at Woods Hole, Mass., August 5-6, 1954

Edited by

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of

The Johns Hopkins University
Baltimore, Maryland

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### Summary.

From the discussions at Woods Hole on August 6, 1954, it was clear that recommendations regarding sea disposal of radioactive wastes could not be made. The uncertainty regarding safety of sea disposal stems from the fact that the physical, chemical and biochemical behavior of the oceans cannot satisfactorily be predicted. Rates of circulation, diffusivity and turnover in the oceans have not been firmly established.

There are many technical aspects to be explored even before experiments can be designed to evaluate the possibilities of sea disposal. The greatest drawbacks appear to be: (a) the lack of suitable electric conductor-cables to go deep in the ocean and (b) the lack of suitable instruments to measure low levels of activity under water.

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# Table of Contents

	Page
Notes on Fission Product Wastes from Proposed Power Reactors by Floyd L. Culler	1
Clay Adsorption of High Level Wastes By L. P. Hatch	32
Radioactive Wastes in a Power Program  By E. G. Struxness	35
Radioactive Wastes at the Savannah River Plant By Joseph Clark	40
Disposal of Low Level Radioactive Wastes By James M. Morgan, Jr	43
OPEN DISCUSSION	
Health Aspects, etc	47

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### OPEN DISCUSSION

	Page
Health Aspects of Radioactive Waste Disposal	47
Work of the NCRP and Others	49
Hazards of Ocean Disposal R. I. Concentration by Marine Organisms Eliminating Hazards of Ocean Disposal Possible Tolerance Levels for Ocean Disposal	
Need for Oceanographic Research	53
Stability and Circulation of the Oceans	54
Circulation Rate in the North Atlantic	55
Dissolved Oxygen Concentration Method to Determine	
Discrepancies between C <sup>14</sup> and O <sub>2</sub> Turnover Rates Biological Interpretation of O <sub>2</sub> and C <sup>14</sup> Turnover Rates	
Turnover Rate from Phosphorus Cycle - Redfield Hypothesis	
BULK DISPOSAL	
Ecological Cycle as Limiting Factor	61 61
Practical Experience with Acid Iron Wastes	62
Disposal as an Experiment	63
Possibility of Sargasso Sea for Bulk Disposal	64
Radioactive Tracer Experiment Possibilities	65
Radiation Measurement Difficulties in Bottom Waters	
Energy Absorption and Upwelling	
Difficulty of Sampling	
Adaptable Instruments Probing Problem	
Cable Problem	
By-passing Cable-Conductor Problem	
Mid-Depth Experiment	
Convection Complications in Surface Waters	
Place and Time of Year for Experiments	

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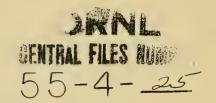
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Instrumentation for Backgrounds	71
Fluorocein Dye Experiment Possibilities	72
Other Diffusion Experiment Possibilities Diffusion Experiment with Drift Bottles	73
DISPOSAL IN SEDIMENTS	
Containing Packaged Wastes in Sea Bottom	75
Natural Adsorption on Sediments	75
Summary	78
Key to Speakers	79

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# Notes on Fission Product Wastes from Proposed Power Reactors

F. L. Culler, Jr., Director Chemical Technology Division Oak Ridge National Laboratory

Date: March 4, 1955



# Distribution

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# Introduction to Nuclear Reactor Terminology.

Types of Fissionable Materials: There are three known fissionable isotopes known which can be used to support a chain reaction in nuclear reactors. These are:  $U^{235}$ ,  $U^{233}$  and  $Pu^{239}$ . Only  $U^{235}$  exists in nature. It occurs as an isotope of the element uranium in an abundance of 0.71%. The others,  $U^{233}$  and  $Pu^{239}$ , are produced by transmutations that follow neutron capture in the parent materials  $Th^{232}$  and  $U^{238}$  respectively. These latter two are known as fertile materials.

The Fission Event: The fission event occurs with a certain probability function after a neutron is adsorbed in the nucleus of a fissionable material. The fissionable material then splits into approximately two equal fragments known as the <u>fission products</u>. Energy is released as kinetic energy of fission fragments, high energy neutrons, beta particles and penetrating electro-magnetic radiation. Most importantly, more than two neutrons are released in this splitting process.

Neutrons, which have no charge, can penetrate the nucleus of a fissionable atom to produce an unstable state which causes fission. One of the neutrons produced in each fission event goes to produce another fission, thus continuing the chain reaction. The other one-plus neutrons are available for capture by inactive reactor components and by a fertile material such as  ${\rm Th}^{232}$  or  ${\rm U}^{238}$  to produce the other fissionable materials  ${\rm Pu}^{239}$  and  ${\rm U}^{233}$  in the following manner:

$$U^{238} + n \rightarrow U^{239} \xrightarrow{\beta} Np^{239} \xrightarrow{\beta} Pu^{239}$$
 $Th^{232} + n \rightarrow Th^{233} \xrightarrow{\beta} Pa^{233} \xrightarrow{\beta} U^{233}$ 

If more than one neutron can be captured by a fertile material for each fission event, more fissionable material can be produced than is consumed in fission. This desirable circumstance is known as breeding.

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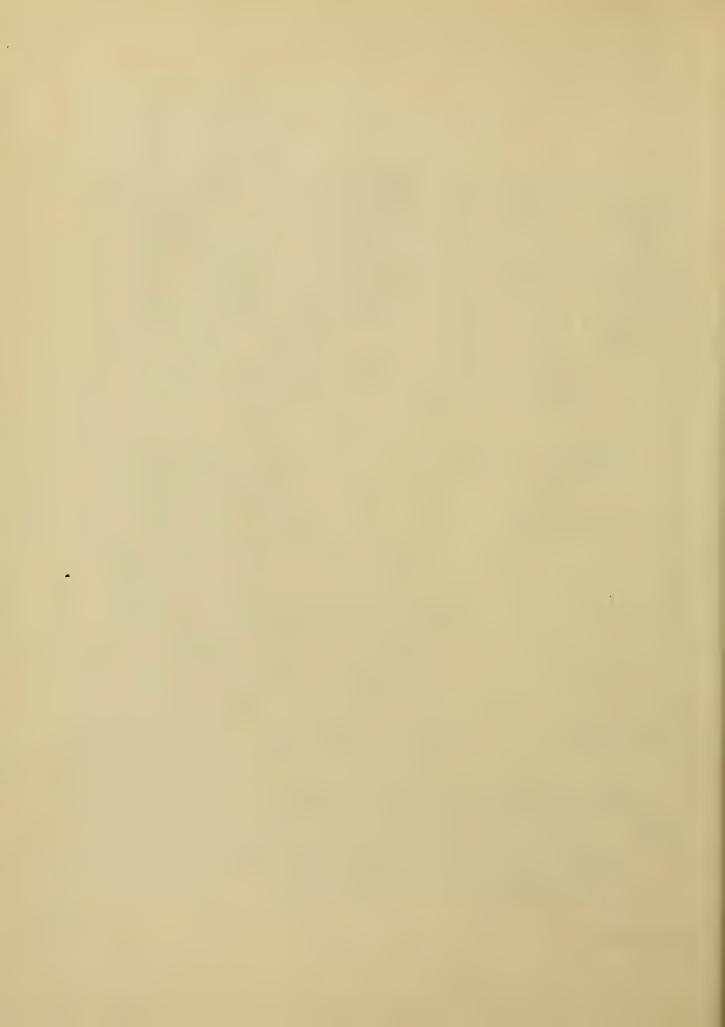


The fission products are of significance in the fission event for three reasons: (1) they are the recipients of most of the energy produced in fission by virtue of the kinetic energy imparted to them at the fission event (energy which is dissipated as heat); (2) fission products have probability for capture of neutrons so that they must ultimately be removed from the fuel, and (3) because many of them, unstable at birth, seek a stable nuclear configuration by the emission of beta particles and radiant energy called gamma rays. The distribution and yield of isotopes produced by fission is well known: isotopes of mass numbers 72 to 162 are produced with individual yields varying from 6% to less than 0.0001% of the total.

Moderators: The fission of U<sup>235</sup>, Pu<sup>239</sup>, or U<sup>233</sup> can be used by man in two ways: (1) as a weapon, (2) as a controlled, chain reacting "pile" for the production of useful power. The fission process yields neutrons with energy much greater than the "rest" or "thermal" energy of the fissionable or fertile nucleus. Fission can be produced by either "fast" neutrons or "slow" (thermal) neutrons, but to date most reactors, except the Experimental Breeder Reactor, have used slow neutrons. The energetic fission neutrons must be slowed down to match the thermal energy of the bombarded nucleus. Because a neutron has no charge, its energy is dissipated best by colliding, usually many times, with other particles or atoms approximately the same size as the neutron. Materials which have a low probability of parasitic neutron capture must be used for this purpose to preserve neutrons for fission or for useful capture. The light elements such as H, deuterium, carbon, aluminum, and beryllium possess these properties.

Nuclear Reactor Principle: A nuclear reactor or "pile" is an assembly of moderator, fissionable materials, coolant, and in some cases, a fertile material arranged in such a geometry that will allow the production of a chain reaction. The fissionable material, or fuel, must be contained in some way to prevent the escape of fission products.

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Need for Processing: Fuel is removed from the reactor before all of the fissionable material is consumed for several reasons: the neutron absorbing fission products build up; radiation damage to fuel elements increases proportionally with time; fuel elements become corroded by reactor coolants; the heat generated causes distortion; and the quantity of fissionable material decreases to the point where a chain reaction cannot be supported. Unreacted fissionable material must be separated from the fission products. Similarly, it is necessary to recover fertile material and the new fissionable material produced by excess neutron capture from reactor systems where fertile material is present. Obviously, fissionable material produced in the fertile material is fissioned along with the fuel but, fortunately, not as fast as it is produced. However, after irradiation, the fertile material contains fission products which must be separated from the unused fertile material and from the new fissionable material.

Cooling Time: After irradiation in a pile, the contaminated mixture of fuel and fertile material normally is allowed to stand for a period of time (cooling period) to allow the fission products to decay and to permit the buildup of new fissionable material from the predecessors, Np<sup>239</sup> and Pa<sup>233</sup>. The fuel elements at the time of discharge are exceptionally radioactive and are too "hot" to be processed conveniently.

Decay of Fission Products: Fission products decay from the instant they are produced in a pile. They decay by emitting beta particles and gamma rays. After 100 days cooling approximately twenty fission products remain which are significant from a waste standpoint. Each one has a characteristic and unchangeable decay rate which is customarily expressed as the half life of the isotope. The decay constant,  $\lambda$ , is related to half life  $T_{1/2}$ , by the following simple expression:

$$\lambda = \frac{0.693}{T_{1/2}}$$
 where: 0.693 = 1n 2



# TOTAL CURIES FOR OPERATION TA LEVEL

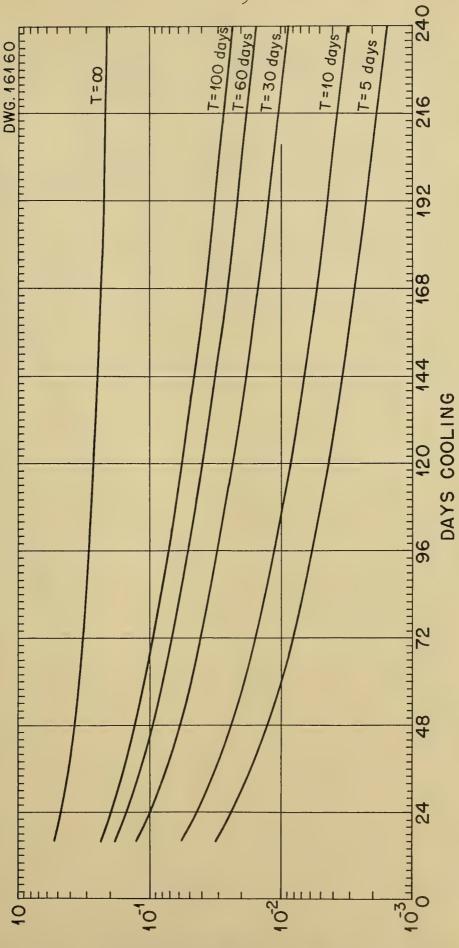
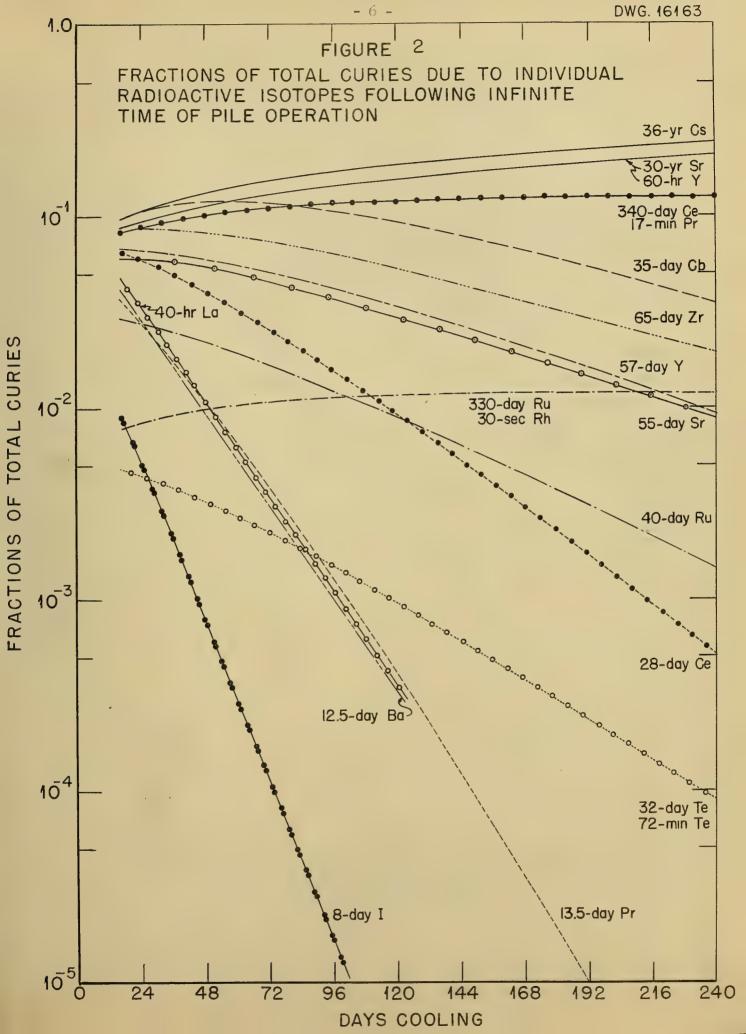
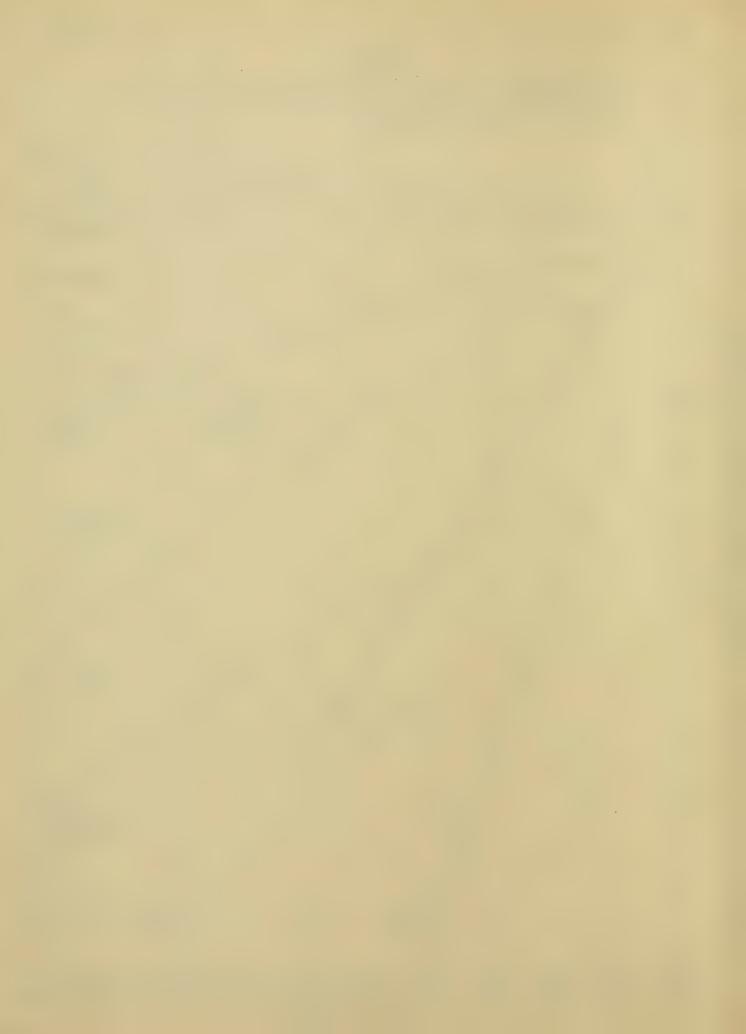
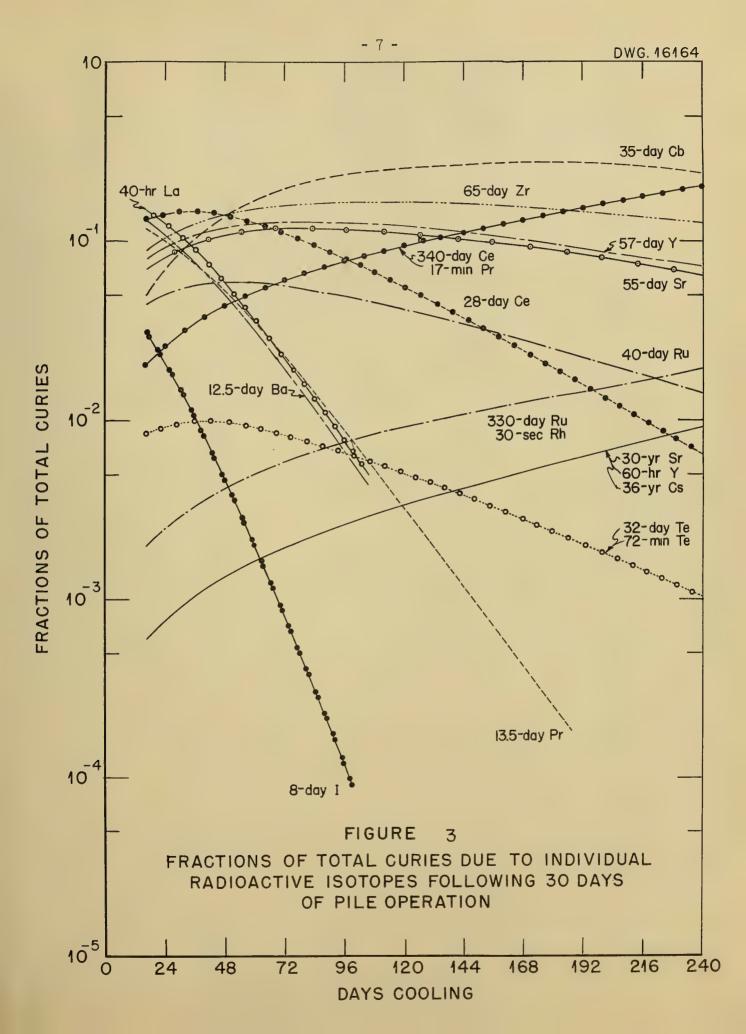


FIGURE 1 TOTAL CURIES IN NATURAL URANIUM (T = TIME OF PILE OPERATION)











# TOTAL WATTS FOR OPERATION TA LEVEL

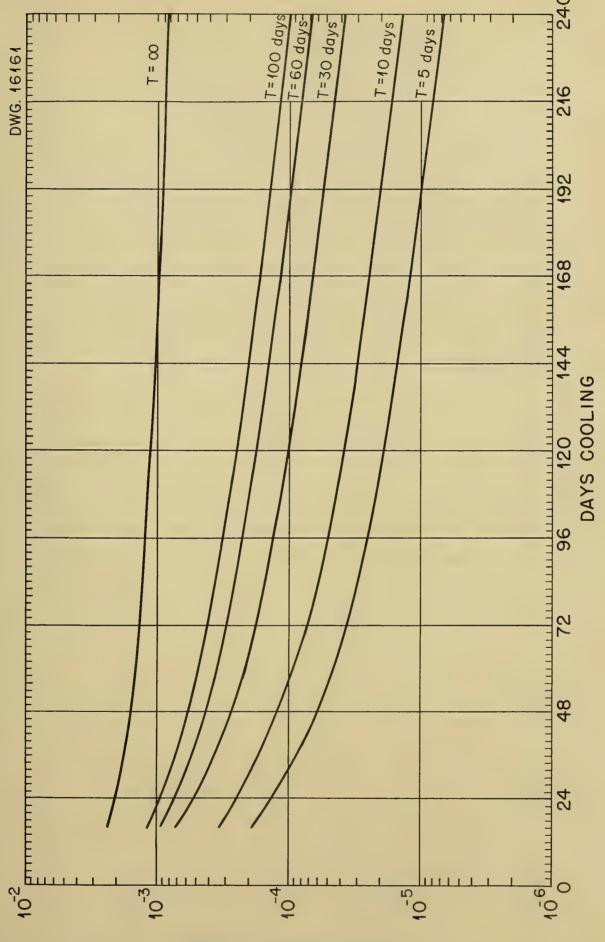


FIGURE 4
TOTAL WATTS IN NATURAL URANIUM
( T=TIME OF PILE OPERATION)



# TOTAL GAMMA WATTS FOR OPERATION LEVEL

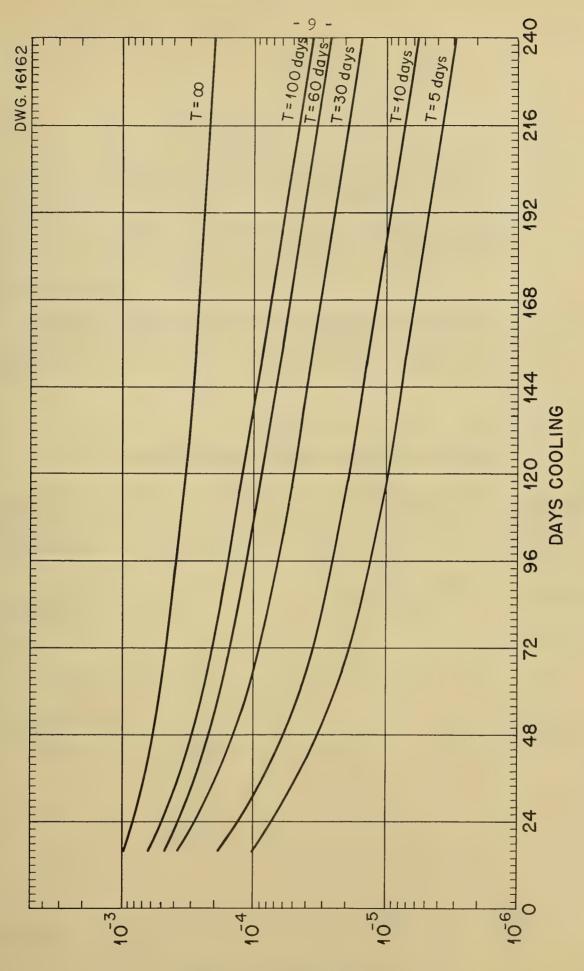


FIGURE 5
TOTAL GAMMA WATTS IN NATURAL URANIUM
( T = TIME OF PILE OPERATION )



Some fission products possess half lives of seconds or less, while others have half lives greater than a million years. Irradiated fuel elements containing the entire spectrum of fission products decay from time of discharge in a manner illustrated by the curves in Figures 1 through 3. Heat released from fission products is given in Figures 4 and 5.

### Types of Reactors

<u>Purpose</u>: Nuclear reactors can be classed first as to purpose. For each purpose there is a corresponding fuel and blanket material as shown in Table 1.

. TABLE 1
Classifications of Reactors According to Purpose

Purpose	Fuel	Fertile Material	New Fuel
Pu production	Natural H (0.71% U <sup>235</sup> )	U <sup>238</sup> (In natural U)	Pu <sup>239</sup>
High flux experimental	v <sup>235</sup>	Optional	Optional
Mobile power	<sub>U</sub> 235	None	None
U <sup>233</sup> production (breeder)	U <sup>235</sup> Full or partial	Th <sup>232</sup>	<sub>U</sub> 233
Commercial power			
a. Thermal	Natural U U <sup>237</sup> U <sup>233</sup>	Th <sup>232</sup>	<sub>U</sub> 233
b. Fast	u <sup>235</sup> Pu <sup>239</sup> U <sup>2</sup> 33	<sub>U</sub> 238	Pu <sup>239</sup>

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Fuel Arrangement: A second distinguishing feature is the fuel arrangement. The original and most developed type is the heterogeneous reactor in which fuel, moderator, coolant and fertile material are separated. A second type is the homogeneous reactors; in this system fissionable and fertile material are intimately mixed with a coolant and a moderator and circulated through a geometrical shape that will sustain a chain reaction in one section of a loop while removing heat with a secondary coolant in another section.

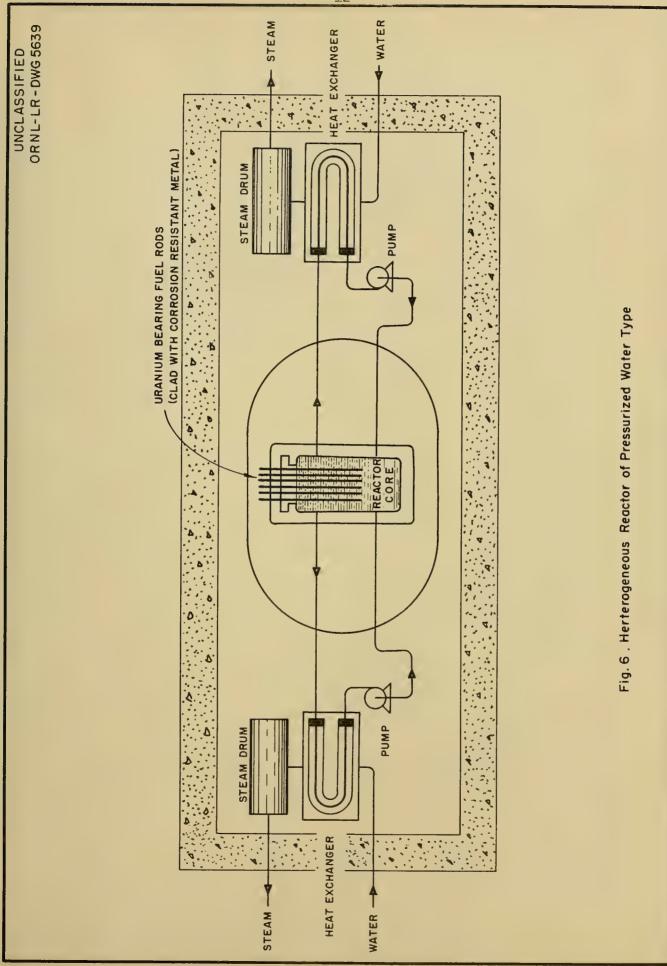
Figure 6 illustrates a heterogeneous natural uranium graphite-water moderated, water cooled reactor. Figure 7 shows an aqueous homogeneous reactor which uses U<sup>235</sup> fuel, has a thorium blanket and is heavy water moderated and cooled. The natural U reactor in Figure 2 illustrates the type of reactor used for Pu production and can be considered as the principle producer to date of fission product wastes.

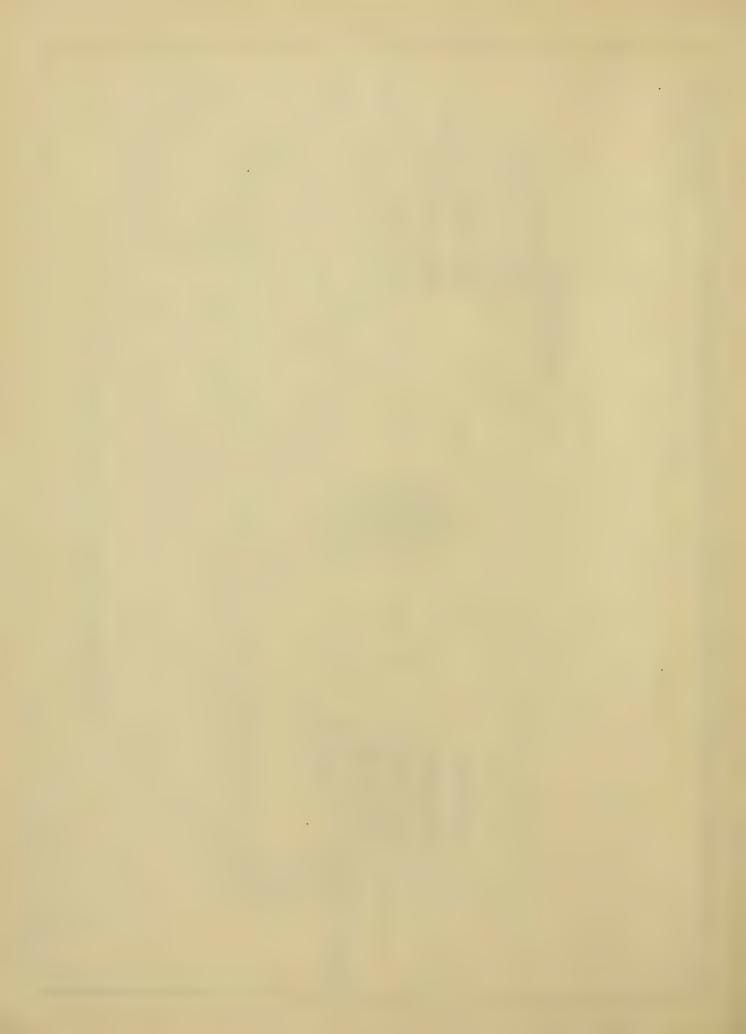
Heterogeneous reactors for mobile power, which use U<sup>235</sup> as fuel, usually require a fuel element in which a metallic diluent is added to the uranium to provide adequate surface for heat removal. As will be shown later, this incactive diluent plus a cladding material add large quantities of salts to waste solutions, thus materially increasing their volumes.

# Relation of Fission Products to Reactor Power

Every time an atom of  $U^{235}$  fissions about 193 mev of energy are released. Of this only 185 mev appears as sensible heat in a reactor. The principle energy is given off as kinetic energy of the fission products. The energy produced in the fission process can be related to the fission products by defining the decay energy after discharge from the reactor. Using  $U^{235}$  as the fuel, a theoretical calculation can be made in the following manner:







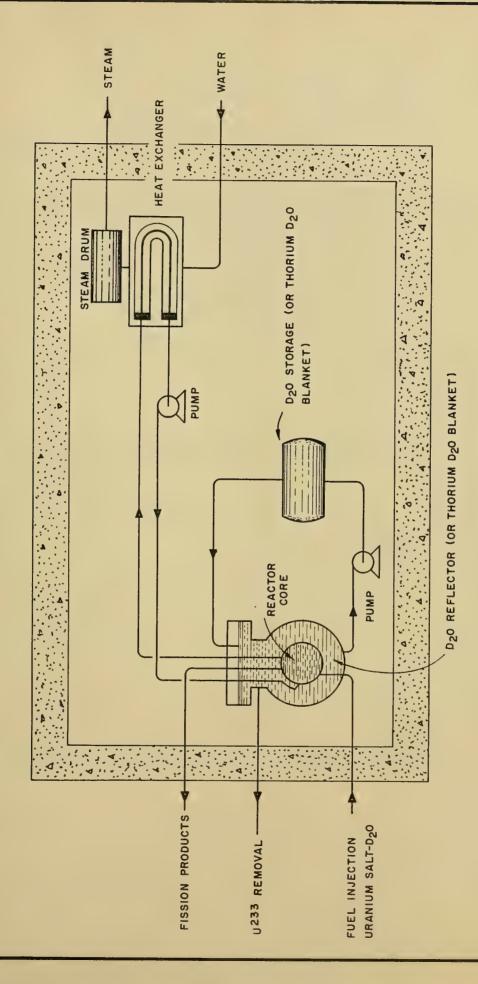
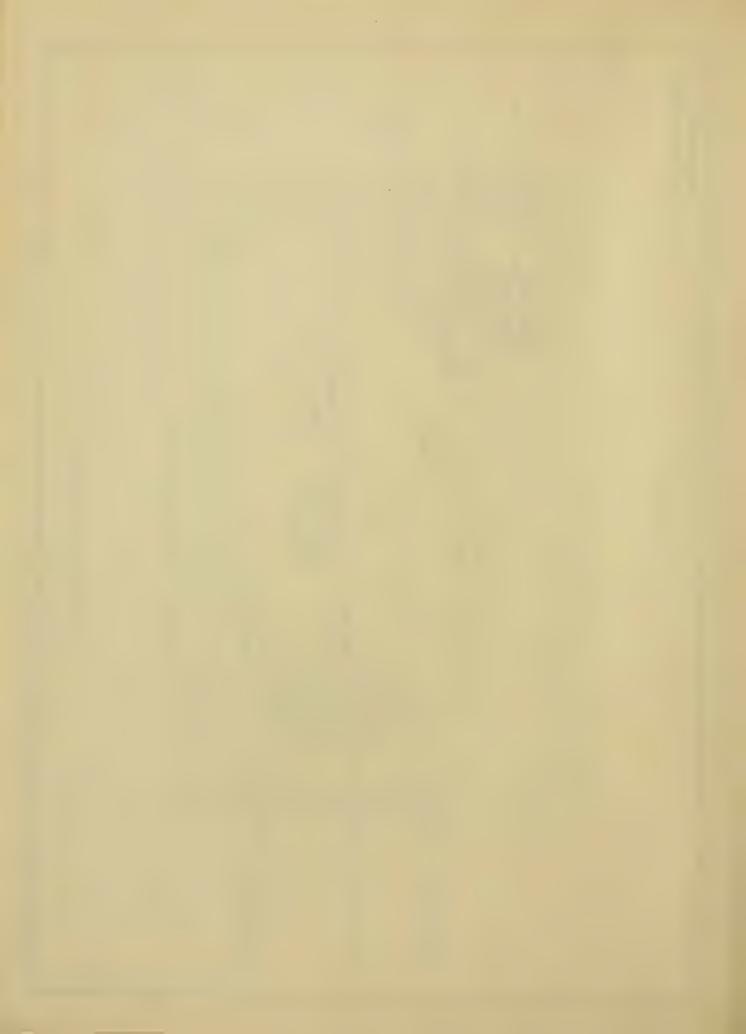


Fig. 7. Homogeneous Reactor



The energy released by fissioning 1000 grams of  $U^{235}$  is:

or 22,800 megawatt hrs

or 1000 megawatt days

This energy of fission is released in several ways as shown in Table 2:

TABLE 2

Energy Distribution from Fissioning 1 Kg U<sup>235</sup>

			Mend
	Mev/fission	%	Mwd 1 kg U235
KE of fission fragments	162	84.0	840
Fission gamma	5	2.6	26
KE of neutrons	5	2.6	26
Weutron capture gamma	~10	5.1	51
Beta decay energy	5	2.6	26
Gamma decay energy	6 193	3.1	31

Note: 1 kg U<sup>235</sup> is contained in 139 kg of natural uranium.



## Magnitude of the Waste Problem - A Hypothetical Reactor.

To define the magnitude of the waste problem, let us consider a reactor in which the following occurs: Enriched uranium fuel is used in a reactor designed to produce 1000 megawatts of heat. If the specific power is approximately 10 kilowatts per gram of U<sup>235</sup> as in the Materials Testing Reactor, the reactor would have to contain 1000 kg of U<sup>235</sup>. If the reactor is operated until 30% of the fuel is consumed, the quantity of fission products produced is approximately 254 grams per kilogram of U<sup>235</sup> initially charged to the reactor. If the fuel is allowed to decay for approximately 100-days after discharge from the reactor, the specific fission products shown in Tables 3 and 4 will be present per 1000 grams charged to the reactor. Table 3 lists those fission products with half lives less than 1 year; Table 4 those with half lives greater than one year.

In the tables curies are listed as beta curies and gamma curies merely to distinguish between beta emissions and gamma emissions. Barium 140 has a fission yield of approximately 6% and dictates shielding at short cooling times. Lanthanum, its decay daughter emits a 2.1 mev gamma, the most energetic of active fission products which exist after short cooling. This γ photon controls shielding at about 60 days cooling. With continued operation of nuclear reactors the longer half lived isotopes  $^{137}$ ,  $^{90}$ ,  $^{114}$  and  $^{129}$  will build up to large equilibrium concentrations as reactor irradiation is continued. Similarly, because of their long half lives, they will be the principle activities that reach large equilibrium quantities for storage or disposal as radioactive wastes.



Table 3

Short-Lived F.P.'s per 1000 gm U<sup>235</sup> Reactor Charge at 100 Days Cooling

30% Burnup

F.P.	Half Life	Grams	Beta Curies	Gamma Curies
Y-90	62 h	4.63	748	-
Rh-106	30 s	0	0	515
Ce-144	275 đ	4.90	16,332	80
Ze-95	65 a	1.52	32,647	<b>62,</b> 356
Nb-95	35 d	1.61	63,657	65,657
Y-91	. 57 d	1.11	28,239	-
Sr-89	55 đ	0.86	23,253	-
Ru-103	45 d	0.46	13,236	6,618
Ce-141	28 d	0.45	15,006	15,006
Ba-137	2.6 m	<b>400</b> 3	-	508
Ru-106	290.0 d	0.35	1,514	-
Pr-143	13.8 d	0.02	1,465	•
Ba-140	12.5 d	0.02	1,222	305
La-140	1.7 d	200	1,222	1,331
I-131	8.0 d	60	23	29
	Inactive			
	Total	15.93	198,564	152,325



Table 4

Long-Lived F.P.'s per 1000 gm U<sup>235</sup> Reactor Charge at 100 Days Cooling

with 30% Burnup

F.P.	Half Life	Grams	Beta Curies	Gamma Curies
			<b>"</b> (0	
Cs-137	. 33 у	7.05	563	-
Sr-90	25 y	4.63	748	-
Pr-144	17 m	4.90	16,333	17,966
Te-129	72 m	0.03	1,217	2,435
		16.61	18,861	20,401
	Inactive F.P.	230.00		
	Short T 1/2	15.93	198,564	152,325
	Total	262.54	217,425	172,726



<u>Wastes Depend on Reactor System</u>: In order to define the wastes more specifically, it is necessary to assume a particular reactor system with its associated chemical plant. Following are two different assumptions:

# Case 1. Natural Uranium:

The reactor contains the 100 kg of  $U^{235}$  in natural uranium (mixture of  $U^{238}$  and  $U^{235}$ ). Such a reactor would produce  $Pu^{239}$ . Assume that 1 gram of Pu is produced per gram of  $U^{235}$  burned. The irradiated fuel from such a reactor would then contain the following after approximately 30% burnup:

	Per 1 kg U <sup>235</sup> Charged or 139 kg natural U charged	Per gram U <sup>235</sup> Consumed, fission + capture
Total U	~ 138.4 kg	_ 465 gm/gm
Pu	$\sim$ 300 grams	∠ 1 gram/gm
FP, inactive + active	~ 265 grams	~0.88 gram/gm
*FP active	_32.5 grams	~0.11 gram/gm
*FP active β	$\sim$ 220,000 curies	_735 curies/gm
* 100 days cooling		

\* 100 days cooling

## Case 2. Non-breeding Reactor:

The reactor uses enriched U<sup>235</sup> as fuel with no fertile material. Here the irradiated fuel would contain the following after 30%: burnup:



	Per 1 kg U <sup>235</sup> charged	Per gram U <sup>235</sup> consumed, fission + capture
Total U	700 grams	~ 2.3 grams/gram
FP active + inactive -	254 grams	$\sim$ 0.85 gram/gram
*FP active	$\sim$ 31.5 grams	~0.10 gram/gram
*FP active β	~ 210,000 curies	~700 curies/gram

\* 100 days cooling

#### Chemical Processing:

Until the present time, most reactors have been of the heterogeneous type. This means that the fuel elements are usually metallic and require complete dissolution in order to be processed. Nitric acid has been the solvent in almost every case. Fission products must be separated from the uranium to the same level as the natural background of uranium; in numbers the separation factors are greater than one part per million.

It is also necessary to separate Th from U<sup>233</sup>, or Pu from U, to an equivalent decontamination factor. Chemical separations are accomplished by completely remote processes; three feet of concrete or more shield personnel. The processing equipment is designed and built to be maintained remotely or be chemically decontaminated before maintenance. Solvent extraction processes have been developed to decontaminate uranium and plutonium to natural backgrounds in two or more cycles.



Processing Methods: Separation of uranium or plutonium from fission products can be accomplished by many methods, the most versatile of which is solvent extraction. Uranium and plutonium can be selectively extracted from an aqueous solution by organic solvents such as diethyl ether and tributyl phosphate. Important steps in a typical solvent extraction process are:

- 1. <u>Dissolution</u> of the irradiated reactor fuel element in a strong acid such as nitric acid. Cladding and alloying elements may be dissolved along with the fuel material, or the cladding dissolved separately and discarded as a slightly active waste solution. Alloying materials usually are dissolved along with the fissionable material, thus contributing materially to the total salt content of waste solutions.
- 2. Feed clarification followed by feed adjustment to flowsheet concentrations. If feed clarification is required, waste solutions containing varying quantities of fission products result from discharged filter or centrifuge flush solutions. Solids result from impurities in fissionable, fertile or cladding material.
- 3. Solvent extraction of uranium and plutonium in highly oxidized, solvent soluble state into the organic solvent. Solvents are highly selective for uranium and plutonium, leaving fission products, cladding material, and alloying diluents behind in the aqueous phase.

In solvent extraction from nitric acid solutions, plutonium and uranium extract as nitrate complexes into the organic phase so the aqueous phase must be "salted" with nitrates. In some processes HNO3 is sufficient; others require "solid" salting agents such as dissolved aluminum nitrate. The organic stream bearing almost all uranium and plutonium originally in the aqueous phase is "scrubbed" with nitrate bearing aqueous phase, which removes additional fission



products from the organic stream and at the same time refluxes some uranium and plutonium to the extraction section of the tower.

The organic stream from the compound extraction-scrub tower passes to a partitioning tower in which uranium and plutonium are separated quantitatively. This is accomplished by reducing the plutonium to a non-extractable valence state using a selective reducing agent for plutonium dissolved in an aqueous phase. Plutonium nitrate is extracted from the organic phase to the aqueous phase, leaving uranium in the organic phase. Uranium bearing organic phase then passes to a stripping tower where the uranium is extracted by water from the organic stream. Several additional cycles of extraction may be required to reduce the fission product activity carried with both the uranium and plutonium streams to acceptable tolerance levels. Fission products are removed by factors greater than one part per million parts of uranium or plutonium; very high recovery of uranium and plutonium are obtained.

- 4. Fission product bearing waste streams are produced from the first column in each solvent extraction cycle, but the first cycle extraction tower raffinates contain over 99.9% of the fission products. This stream may be combined with others to constitute the radioactive wastes from a chemical reprocessing plant.
- 5. Radioactive wastes from other sources contribute to the overall waste picture. Vessel off-gases, and particularly dissolvers, may be scrubbed to remove fission product gases such as I<sup>131</sup> and entrained radioactive spray and solids. The organic solvent used in the process is recycled for reuse only after a cleaning operation which produces slightly active wastes.



The solvent extraction processes may be grouped into three general categories for the purpose of describing the characteristics of the wastes which they produce.

A salted process uses a solvent which favors the formation of organic soluble nitrate like ether. It is necessary to add an inorganic complex of uranium and plutonium. The salting agent, such as aluminum nitrate or copper nitrate, is contained in the waste and is the principal chemical to be dealt with in the disposal problem. The <u>nitric acid salted system</u> uses another solvent, tributyl phosphate, diluted with a hydrocarbon diluent. This system has the advantage of requiring no salting agent other than nitric acid so that the wastes contain few inactive salts since the nitric acid can be removed by evaporation. The <u>enriched U<sup>235</sup> diluent salted process</u> is quite similar to the salted natural uranium-plutonium process, but in this the salting agent may be the alloying element in the fuel assembly. The diluent follows fission products into the highly radioactive first cycle waste stream.

Physical Properties of Wastes: The physical properties of the wastes resulting from the processes described are listed in Table 5. The solid salted process waste may be concentrated by a factor of 3 by evaporation, but this is the limit for the residue is then solidly set-up. The volume of solids in some of these wastes after evaporation may be reduced by only a factor of 2 because of bulking.

Wastes may be stored as they come from the extraction tower, but some are neutralized, usually with sodium hydroxide, before they are stored. A large volume increase upon neutralization in the enriched U<sup>235</sup> process occurs due to the presence of amphoteric salts which require a large volume of neutralizing agent. The neutralized volume is approximately 4 times the unneutralized volume.



Table 5
Physical Nature of Wastes

	Natu	Enviolated II 025	
Properties	Salted	HNO3 Salted	Enriched U-235 Diluent Salted
BP	102°C	112°C	103°C
Sp. Gr.	1.18	1.24	1.23
Concentration Possible	3	?	2
Gal/gm U-235 Burned (Unneutralized)	2.5	0.5	5.0
Gal/gm U-235 Burned (Neutralized with NaOH Solution)	10.0	1.5 to 2.0	20.0



Chemical Properties of Wastes: Table 6 presents data on the chemical nature of the wastes. In the natural uranium-plutonium salted process and in the enriched  $U^{235}$  process, the wastes may be acid deficient, whereas the nitric acid salted system may be 8 N nitric acid.

Precipitates may form in a relatively short period of time after neutralization of wastes from either of the salted systems shown. Such precipitates as aluminum and iron hydroxides which later convert to oxides, make handling, storage, and disposal of radiochemical wastes more difficult.

Radiochemical Properties of Wastes: The radiochemical nature of wastes is shown in Table 7. These values are approximations for a reactor similar to the Materials Testing Reactor having 10 kw/gm  $U^{235}$  specific power, 30% burnup, 1000 megawatts total heat and a consumption of 1 kg of  $U^{235}$  per day.

The shielding requirements given in Table 7 are based on personnel tolerances of 6.5 milliroentgens per hour. The need for shielding is one of the things that makes shipping both expensive and difficult.

Heat is liberated in wastes by decaying fission products. This heat is estimated in watts per gallon in Table 7. The decay heat after 100 days cooling is sufficient to cause boiling of wastes stored in underground tanks. Boiling causes further complications, one of which may be the potential hazard of the release of particulates to the atmosphere. The heat also magnifies corrosion difficulties. There is always this choice to make: a) store the acidified wastes and cope with the corrosion problem by cooling large tanks to 120-150 °C or, b) neutralize the wastes, allow them to boil, let all of the solids precipitate and then gamble with "burping", or the blowing of solids up into the condensers.



Table 6
Chemical Nature of Wastes

Natural U-Pu			Transched II 025
Properties	Salted	HNO <sub>3</sub> Salted	Enriched U-235 Diluent Salted
Acidity, N	~0.3 (Basic)	8.0	-0.2
Total Salts, M (Unneutralized)	1.2	0.2	2.0
Total Salts, $\underline{M}$ (Neutralized)	6.0	8.2	Acid Storage
Solids Stability	Unstable Basic	Stable	Unstable Basic



Table 7

Radiochemical Nature of Waste

	Natural U-Pu		
Properties	Salted	HNO <sub>3</sub> Salted	Enriched U-235 Diluent Salted
Curies/gal Acid	80	400	2,000
Curies/gal Basic	20	200	Acid Storage
Shielding, Lead 1 cc Acid	3.5"	) <sup>‡</sup> 42	)† 12
Shielding, Lead 500 gal Acid	11.5"	12"	12"
Watts/gal Acid	0.3	1.2	5.4



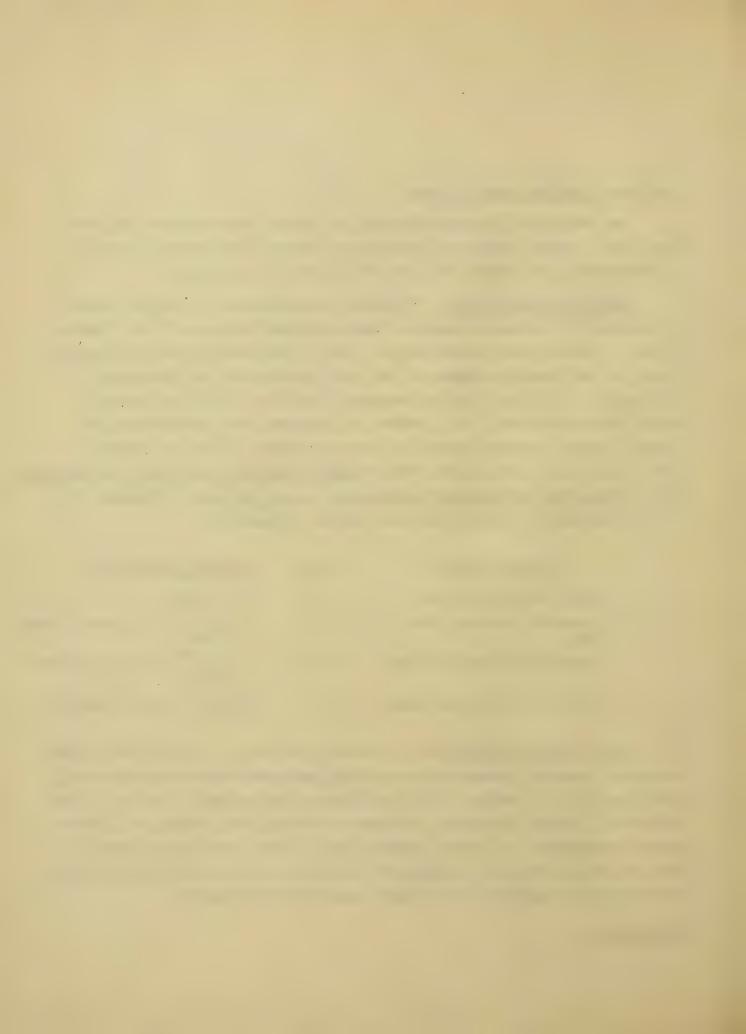
## Problems of Handling Reactor Wastes

The problems of cooling hot wastes and coping with corrosion have been mentioned. Another problem of considerable economic significance is that of transporting the wastes from production site to disposal site.

Health Tolerance Levels: The handling and disposal of fission products is regulated by biological exposure limits or human tolerances to the fission products. These specifications depend upon: a) the chemical and radiochemical nature of the fission products; b) the site in which they are deposited; c) the length of time that they are retained by the body; and d) the method by which they are adsorbed, i.e., whether as dissolved ions, particulates, or gases. General tolerance levels for unknown mixtures of fission products are roughly as follows as taken from, "Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Concentrations in Air and Water", Handbook 52, U. S. Department of Commerce, National Bureau of Standards:

	Kind of Uptake	Type	Maximum Weekly Dose
1.	Total body irradiation	β, γ	0.3 r/week
2.	Ingestion in water, food, etc.	β, γ	3 x 10 <sup>-7</sup> μ c/cc water, based on Sr90
3.	Inhalation of particulates	β, γ	$2 \times 10^{-10} \mu$ c/cc air, based on Sr90
4.	Inhalation of fission gases	β, γ	$3 \times 10^{-9} \mu$ c/cc, based on

Waste Shipping Problems: As is evident from Table 7, which shows the lead shielding required, transport entails moving very much shield with relatively small quantities of wastes. With liquid wastes the problem of handling is very difficult. Leakage or spillage of a very few drops of the wastes will produce enough contamination to prevent shipping, and in many cases prevent getting near shipping containers. Handling the waste as a solid is also very difficult because the dust particles which might escape are very dangerous.



The transportation of radioactive waste solutions is expensive since carriers must be shielded with many inches of lead. Carriers weigh almost as much empty as when full.

#### Current Disposition of Radioactive Wastes

Radioactive wastes are stored in large tanks for indefinite periods of time. Only relatively small quantities of fission product activity have been released to ground or to ground waters. Experimental dumping of three to five year old liquid wastes has been in progress at Oak Ridge National Laboratory for approximately two years. However, no feasible method for "ultimate" disposal of large quantities of liquid wastes has been practiced or proposed, long term storage of wastes in carefully constructed and monitored waste tanks is the current practice. Storage costs, in tank investment alone, are approximately \$1.00 to \$2.00 per gallon.

#### Proposal for Less Expensive Storage Pit

The high cost of waste storage and the lack of a cheap, safe solution to waste disposal, other than storage, has spurred development work on perfecting cheaper storage methods. At Oak Ridge National Laboratory, we are experimenting with earth pits lined with an impermeable material such as asphalt. It is proposed to fill such a pit with sand to a depth of approximately 30 ft, then to fill the pit approximately half full of liquid waste. Because of radioactive decay heat, the liquid wastes will boil, releasing steam through approximately 15 ft of sand filter to the air. The sand above the boiling liquid waste will act as a shield and will prevent entrainment of particulates during self-concentration. Obviously such a storage pit would be located in a restricted access area and possibly would require a rain deflector roof. Costs for storage in such a pit, assuming that a satisfactory liner can be developed and that boiling liquids can be maintained many feet below the surface of the sand, have been estimated to be about \$0.15 per gallon of liquid wastes. The feasibility of this type of pit storage and its hazard potential will



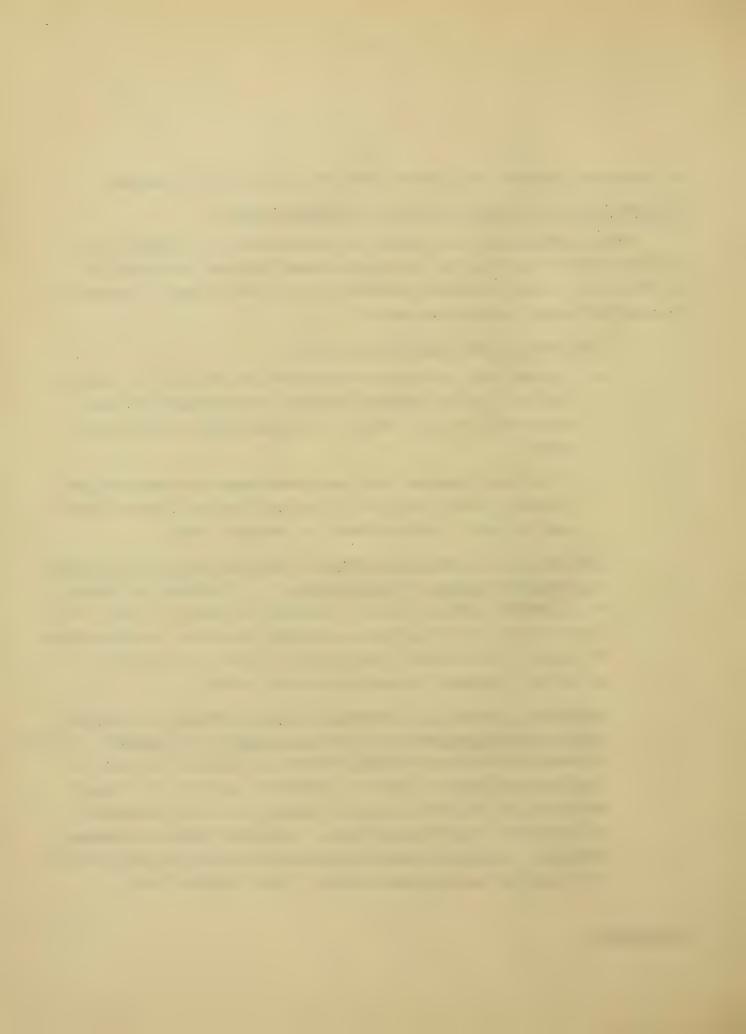
be evaluated. However, it is not an answer for ultimate waste disposal.

# Proposed Methods of Waste Processing for Ultimate Disposal

Many proposals have been studied for concentrating and fixing fission product wastes so that they can be safely released to either the ground or to the ocean. A list of methods considered follows, with comments concerning limitations imposed by safety and costs.

#### 1. Concentration and disposal as a liquid

- a. To deep wells or to salt domes-hazard not evaluated but probably serious. Cost of shipment of wastes to few suitable disposal sites would be high. Hazard of shipping highly active liquids great.
- b. To the sea, using sea water as diluent-hazard not evaluated but obviously great, since the ecology and dilution of entire oceans must be known. Cost and hazard of transport high.
- 2. Evaporation to dryness and disposal of solids in can, cast in concrete, or fused into a glass or similar material evaporation to dryness would present a serious hazard in dusting, cost would be high. Heat evolution in a solid would present problem of cooling. Activity could be leached from solids by ground water or ocean. Cost of transportation to ultimate disposal sites would be high.
- 3. Absorption, adsorption or exchange of fission products on naturally occurring clays, preliminary to firing to a solid or to canning all clays or naturally occurring exchange materials have limited capacity for ionic species involved, none will remove all ions desired. Organic exchangers are subject to radiation damage; also do not permanently fix activity. Inactive salts load or completely block most exchange materials. Exchange or absorption processing facilities would have to be as large as main processing plants. Costs would be high.



4. Removal of specific fission products for use as radiation sources - desirable to provide revenue to cover costs of waste disposal, but does not eliminate the need for an ultimate disposal need.

To summarize, there is no obvious answer to the ultimate disposal of fission products. A most ingenious invention is required, and a large and expensive development program will be necessary to answer the safety problems of ultimate disposal. It is my opinion that there is no very promising method for ultimate disposal.

#### New Reactors and Their Effects on the Wastes

Reactors which differ from those discussed above, will produce wastes of different character. Proposed reactors using difficultly soluble fuel elements such as those containing zirconium or stainless steel, will greatly increase the wastes from aqueous processes. The temperature resistant fuels required for mobile power reactors require large dilutions of uranium with materials such as stainless steel, inconel, or zirconium. Dissolution of these materials in inorganic acids results in high concentrations of inactive salts in the fission product wastes. Corrosive anions such as  $\mathrm{SO}_{\frac{1}{4}}$ ,  $\mathrm{Cl}_{\frac{1}{4}}$  F also may be present. It is possible that such fuels can be reprocessed by utilizing the volatility of UF $_6$  to separate it from fission products. The volume of waste from such a process probably would be small. However, the heat and corrosion problems still would be present.

Reactors of the homogeneous type offers some hope for isolating fission products in small packages. It may be possible to remove precipitated fission products with essentially no diluant from homogeneous reactor fuels. However, problems of ultimate disposal remain unchanged.

#### Approximate Economic Requirements for Waste Disposal in a Power Economy

Assuming that power in the United States should cost approximately 8 mils/kwh to be competitive, assuming that chemical reprocessing should cost not more than

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20% of this, and further assuming that waste disposal should cost not more than 20% of the reprocessing, we can calculate that the cost of waste disposal should be approximately 0.32 mils per kwh of electricity. If the reactor-power plant combination operates at 25% thermal efficiency, 4 kwh of heat must be produced per kwh of electricity. Thus heat must be produced for 2 mils/kwh; the waste disposal cost should be approximately 0.08 mils/kwh of heat. To produce 1 kwh of heat 4.2 x 10<sup>-5</sup> gm U<sup>235</sup> must be burned. Assuming that the process produces 2 gals of waste per gram of U<sup>235</sup> burned, then the cost of waste disposal should not exceed \$1.00 per gallon. This figure may be representative for wastes from a non-salted solvent extraction process. For a salted process, or one where the fuel provides large quantities of dead salts, wastes may be produced at the rate of 5 gal/gm U<sup>235</sup> burned, which would decrease the allowable cost to \$0.40 per gallon. If wastes contain 400 curies per gallon the per curie cost would be 0.25 cents and 0.10 cents per curie.



#### CLAY ADSORPTION OF HIGH LEVEL WASTES

L. P. Hatch, Sanitary Engineer Brookhaven National Laboratory

#### Objectives of Research Project.

To facilitate the handling of reactor wastes it is desirable to separate the radioactive fission products from the main body of the wastes, and to get them in the smallest volume possible. At Brookhaven a research project was undertaken to find an economical way to adsorb and fix fission products on montmorillonite clay. The solution to the problem is not a simple matter of flowing the waste streams through a bed of the clay. The reactor waste streams are too complex for that. The composition of the waste stream depends upon the type of fuel used in the reactor, which in turn depends upon the particular use of the reactor.

#### Composition of Reactor Wastes Depends upon Form of Fuel

Common Form: In the heterogeneous type of reactor the solid fuel elements are made up of uranium containing U<sup>235</sup>. After fissioning in the reactor, fission products are interspersed uniformly throughout the fuel element. To separate uranium and plutonium from fission products, the irradiated fuel element is dissolved in nitric acid and the solution put through various solvent extraction processes. After separation the fission products are in the acid waste stream of the chemical processes. The acidity of the wastes causes much trouble in subsequent stages of handling the fission products. In the work at Brookhaven National Laboratory the first problem is to make a separation of the acid or the other inert salts in order that adsorption of the fission products on clay may proceed with efficiency.

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#### Other Fuel Forms

Uranium alloyed. The enriched uranium is combined with an alloying material like aluminum. The dissolution of this fuel element for processing results in a solution containing a high concentration of aluminum nitrate instead of nitric acid.

Aqueous uranium solution. The uranium can be in a water solution in the form of uranyl sulfate or uranyl nitrate. The same solvent extraction processes will be required to separate the uranium from the fission products and presumably acid wastes like those from the heterogeneous reactor would result.

Uranium in metal solution. The uranium fuel can be dissolved in liquid bismuth metal. The bismuth will serve to carry the uranium in solution in a homogeneous form and also to take heat away from the reactor; it is a good heat exchanger. The fission products will then be in a metallic state (with zero charge) and can be removed by the addition of a fused salt instead of by many solvent extractions. Further concentration of the fission products from the fused salt may be possible by an electrolytic procedure.

#### Condensing Fission Products in Montmorillonite Clay.

Natural montmorillonite clay has an ion exchange capacity of about one milliequivalent per gram. (The clay weighs about 100 lbs. per cubic foot.) The concentration (of fission products) attained on clay depends upon the chemical solution the wastes are contained in. Waste solutions containing H ions or Al ions are not favorable because these ions usually outnumber the fission products and will use up the

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exchange capacity before much of the radioisotopes can be adsorbed.

It is necessary to neutralize the acid wastes.

Neutralizing Acid Wastes: To neutralize the acid with NaOH for adsorption defeats the purpose of the process because the Na ion is just as bad as the H ion. Ion permeable and semi-permeable membranes in conjunction with an electric current offer possibilities for bypassing chemical neutralization. The current drives the oppositely charged ions to oppositely charged terminals leaving the solution virtually neutralized. Aluminum nitrate can be treated much the same way.

Activity Concentration in the Clay: As an indication of ultimate activities in the clay, about 45 mg. of pure  $\mathrm{Sr}^{90}$  can be put in 1 gm. of clay. Since there are approximately 200 curies of activity per gm. of  $\mathrm{Sr}^{90}$ , 45 mg. of  $\mathrm{Sr}^{90}$  will amount to roughly 10 curies per gram of clay. If the isotope has a half-life of one year instead of 20 as  $\mathrm{Sr}^{90}$  has, and if the same amount of the isotope can be put on the clay, the initial activity will be 20 times as great as would be the case with  $\mathrm{Sr}^{90}$ . With mixed fission products it is estimated that about 10 curies of aged material can be adsorbed per gram of clay.

Ruthenium causes trouble in the clay adsorption process by acting as an anion. It will pass through the cation exchanger, clay, but can be later collected on an anion exchanger.

By firing the clay to about 800° C., the structure of the clay is altered and the adsorbed fission products become permanently attached. The resulting product is practically unleachable. Leaching from the flay in a flowing solution levels off with time and amounts to about 0.2%.

(For a further discussion of the clay adsorption process, -34see Dec. 1954, "Nucleonics".

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#### RADIOACTIVE WASTES IN A POWER PROGRAM\*

#### E. G. Struxness - Health Physics Division Oak Ridge National Laboratory

#### Electric Power Expansion in the United States.

To arrive at an estimate of the quantity of waste radioactivity which may be of concern to us in the future, a logical idea would be to incorporate power reactors in the electrical industry's future expansion. Putnam, Briggs, Starr, Thompson and others have estimated future electric power requirements of the United States. An orderly, constant increase has been predicted to the year 2000 A.D., of about 6.5 to 9 x 10 km generating capacity.\*\*

Power Reactors in the Program: it is necessary to make certain assumptions to fit power reactors in the electric power program. First, assume that the reactors will generate about 10% of the power increase. Next, assume that the reactors will provide 1000 megawatts of heat per day with about 450 megawatts available for electric power. There will be 10 reactors to a power station and about 200 stations around the country.

The hypothetical reactors are of the aqueous, homogeneous, thorium breeder type having its fuel circulating constantly and constantly processed to remove fission products. Of the 450 megawatts

<sup>\*</sup>Based on a paper by Emerson, Straub and Struxness of the Oak Ridge National Laboratory, Dated July 15, 1954.

<sup>\*\*</sup>This is a 6500 to 9000% increase in the 1954 installed generating capacity in the U.S. It is the opinion of the Johns Hopkins group that this estimate is too high by at least one order of magnitude. If the U.S. population doubles by the year 2000 A.D. and if the per capita consumption of electricity trebles during this period, the increase would be about 600% in KWH consumed. Estimates made by the Hopkins group are, therefore, about one tenth those presented here.

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available power, 320 megawatts come from the core and 130 megawatts of thermal power from the thorium blanket. The core will be processed at the rate of 2% per day, which gives, on the average, a 30 day cooling period. The blanket will be processed at the rate of 0.5% per day, providing a cooling period of about 180 days.

#### Fission Product Wastes

Wastes from Individual Reactor: To get an idea of the total amount of radioactivity produced by each reactor over a period of years it is assumed that the wastes from each power reactor are accumulated in a localized volume.

Table 1 lists the significant fission products, including daughter products, produced daily in processing the irradiated fuel and blanket. Chains of the daughter products have been considered as having 3 members; some are actually longer. Gaseous fission products have been disregarded as have isotopes less active than 10 curies and radioactivity produced by neutron capture. In the table, calculated activities are listed for these fission products in the first 4 columns. The 3 right hand columns show the calculated equilibrium levels at the end of 10 years, based on the processing plant output accumulating day by day for 10 years. The total equilibrium activity is about 625 megacuries (625,000,000 curies) from each reactor.

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Table 1. Radioactive Output from One Reactor

Activity in Disposal Pit Daily Output of Chemical (Mega Curies) Processing Plant (curies/day) 10 Year Equilibrium Nucleii Tota1 Core Blanket Core Blanket Total Sr89 0.16 5.23 66,400 2,060 68,460 5.07 Sr90 1,290 596 1,886 13.5 6.24 19.74 y90 1,350 1,946 13.5 6.24 19.74 596 y91 84,000 0.21 7.10 2,580 86,580 6.89 Zr95 84,600 3,340 87,940 79.0 3.12 82.12 Nb95 168.52 162,000 7.72 6,920 168,920 160.8 Ru 103 3.83 178 63,178 3.82 0.01 63,000 Ru 106 512 5,132 2.43 0.27 2.70 4,620 in equilibrium with Ru 106 Rh 106 Ag 111 0.01 607 0.01 607 Cd115 14.7 14.6 0.3 ----Sn 123 13.6 11.7 1.93 Sn 117 91.5 0.01 91.5 Sn 125 62.7 \_\_ 62.7 ---Sb 125 0.13 87.4 0.04 25.1 0.09 62.3 Te 125 0.04 576.6 0.17 0.13 539 37.6 Sb 127 17.8 17.8 -----Te 127 (isomer) 15.0 --15.0 Te 127 0.01 0.03 70 192 0.02 122 Te 129 5,580 32.8 5,613 0.26 0.26 (+ isomer) 9,830 9,830 0.11 0.11 Te 132 670 670 -in equilibrium with  $Te^{132}$ I 132 Cs 136 52.6 52.6 --Cs 137 794 13.8 45.5 1,820 2,614 31.7 Ba 137 in equilibrium with Cs 137 Ba 140 8.89 8.89 48,100 1.6 48,100 La 140 10.23 55,600 10.23 - --55,600 1.8 Ce 141 3.59 82,500 82,865 3.57 0.02 365 Pr 143 0.91 45,900 2.1 45,900 0.91 ---Ce 144 33,410 11.75 1.51 13.26 29,600 3,810 Pr 144 in equilibrium with Ce 144 Nd 147 2.54 2.54 0.1 16,000 16,000 147 136.54 29.5 166.04 Pm 6,350 1,400 7,750 155 Eu 18.3 93.1 0.08 0.08 74.8 156 126 Eu 126 836,500 540 85 625 Total 808,000 28,500

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Wastes from Reactor Program at 2000 A.D.: From the 200 nuclear power stations, 2000 times the volume and activity produced in a single reactor is expected at the year 2000. The total volume would be of the order of a million gallons per day with about a ton per day of fission products.

Radiation Associated with Wastes: The potential hazard associated with nuclear reactor wastes is very real. For a concept of the magnitude, assume that the wastes from a single reactor, accumulated over 10 years, were spread uniformly over 1000 square miles. The ionizing radiation, at a height of 4 feet, is about 4.8 roentgens per hour. The permissible radiation dose is but 300 milli-roentgens per week or 0.0075 roentgens per hour.

If the wastes were kept in the pit, heat generated by the radioactivity becomes a very serious problem unless it is dissipated by heat exchangers or some other device. Making some liberal assumptions with regard to the pit and with regard to heat losses, theoretical temperature calculations indicate ranges up to 19,000° C.

Ranking of Hazardous Radioisotopes: Table 2 lists the critical radioisotopes, arranged in order of relative hazard. This ranking was determined by dividing the amount of each isotope accumulated over 10 years by the concentrations allowed in water and in air as specified by Handbook 52 of the Bureau of Standards. Sr<sup>90</sup>, in every case, is most critical.

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Table 2. Relative Ranks of Hazardous Radioisotopes

On Basis of Daily Output of On Basis of Activity in Chemical Processing Plant Disposal Pit (curies/day) (mega curies)

Core	Blanket	Total	Core	Blanket	Total	
On basis of ingestion in water (MPC)						
sr <sup>90</sup>	Sr <sup>90</sup>	Sr <sup>90</sup>	sr <sup>90</sup>	Sr <sup>90</sup>	Sr <sup>90</sup>	
Sr <sup>89</sup>	Sr <sup>89</sup>	Sr <sup>89</sup>	Nb <sup>95</sup>	Nb <sup>95</sup>	<b>n</b> ь <sup>95</sup>	
I <sup>131</sup>	Nb95	<sub>1</sub> 131	Sr <sup>89</sup>	Cs <sup>137</sup>	sr <sup>89</sup>	
Nb <sup>95</sup>		Nb95	Ba <sup>140</sup>	S <b>r</b> <sup>89</sup>	Cs <sup>137</sup>	
Ba <sup>140</sup>		Ba <sup>140</sup>	Cs.137		Ba <sup>140</sup>	
			Pm147		Pm147	
			I <sup>131</sup>		I <sup>131</sup>	
On basis o	f air intake (	MPC)				
Sr <sup>90</sup>	Sr <sup>90</sup>	Sr <sup>90</sup>	sr <sup>90</sup>	Sr <sup>90</sup>	sr <sup>90</sup>	
Ce <sup>144</sup>	Ce <sup>144</sup>	Ce <sup>144</sup>	Ce <sup>144</sup>	Ce <sup>144</sup>	Ce <sup>144</sup>	
Sr <sup>89</sup>	Sr <sup>89</sup>	Sr <sup>89</sup>	Pm147	Pm147	Pm <sup>147</sup>	
I <sup>131</sup>	Y <sup>91</sup>	r <sup>131</sup>	Nb95	Sr <sup>89</sup>	Nb <sup>95</sup>	
Y <sup>91</sup>	Nb <sup>95</sup>	Y <sup>91</sup>	Sr <sup>89</sup>	Cs <sup>137</sup>	Sr <sup>89</sup>	
Ba <sup>140</sup>	Ru 106	Ba <sup>140</sup>	<sub>Y</sub> 91	Nb <sup>95</sup>	Cs <sup>137</sup>	
Nb95		Nb <sup>95</sup>	Cs <sup>137</sup>	Y90	Y <sup>91</sup>	
Ce <sup>141</sup>		Ce <sup>144</sup>	Ba140	<sub>Ru</sub> 106	Ba140	
			Ru 106	Y <sup>91</sup>	<sub>Ru</sub> 105	
			I <sup>131</sup>		Y <sup>90</sup>	
					I <sup>131</sup>	

In the case of the critical nuclides determined from the air MPC values, this will be their order of hazard if they are taken into the body through breathing. This presupposes that these nuclides are present in the air breathed. If not present, certainly they will not be hazardous.

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#### RADIOACTIVE WASTES AT THE SAVANNAH RIVER PLANT

Joseph R. Clark - Technical Division E. I. du Pont de Nemours & Company

#### Origin of Radioactive Wastes

All of the important wastes at the Savannah River Plant, except a portion of those from the laboratories, originate in the reactors. Nearly all of this radioactivity which remains after radioactive decay is finally present in the wastes from the separation processes. In addition, there are wastes from metal processing, laundries and laboratory research operations. Handling High Level Wastes

Multi-megacuries aptly describes the amount of radioactivity associated with the high level wastes. These amounts and levels constitute a difficult ultimate waste disposal problem.

The irradiated fuel elements are given a cooling period after they are taken from the reactors to allow handling with the shielding provided in the plant. As the irradiated fuel proceeds through the separation processes the levels of activity decrease rapidly. At the end of the separations process the waste is too voluminous to be economically contained. It must be evaporated.

#### Neutralization and Evaporation

Before or after evaporation, depending on the particular waste stream, the waste is neutralized with caustic before being sent to the waste storage tanks. Large amounts of inert salts are formed in the neutralization. The fission products amount to a very small fraction of the final total solids content of the waste.

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Evaporation of the reactor wastes is accomplished in a two stage evaporating system. An over-all decontamination factor of the order of 10<sup>8</sup> is achieved. The waste is evaporated to 35% solids; beyond this point certain materials in the slurry residue would start to crystallize. The residue is then jetted to one of a group of holding tanks.

There are "General Purpose Evaporators" for various low activity wastes, including those from the laboratories. A special evaporator is provided to handle the higher level laboratory wastes.

#### Tank Storage

The evaporator residue is received in 750,000 gallon tanks constructed of steel and concrete. The tanks are equipped with cooling coils to dissipate radioactive decay heat. An air space separates the steel wall from the outer shell of reinforced concrete. The steel tank rests on a saucer-like bottom which would catch and allow detection of any leakage from the tank into the annular air space. All of the tanks are buried underground for effective shielding. Gas filtering devices on the tank vents prevent radioactive contamination of the atmosphere.

Clay 100 to 200 feet thick underlies the storage tank area. Laboratory tests have shown this clay to be a very good ion exchange medium. It therefore provides a factor of safety in case of damage to the tanks. Monitoring wells surround the area, warning of any leakage outward from the storage area.

#### Handling Low Level Wastes

Low level wastes make up a large percentage of the total volume of the Savannah River Plant wastes.

Wastes of activity levels below those which are sent to the General Purpose Evaporators are to be discharged to large earthen basins. In these basins the wastes are dissipated by seepage through the soil, and to a lesser extent, by evaporation. The activity of low level wastes sent to the basins is limited to 10<sup>-10</sup> curies per milliliter. These basins are also underlaid by clay, so a large decontamination factor is anticipated before any radioisotopes reach the water table.

It is the policy of the Savannah River Plant to limit
the discharge of radioactivity to the environment to the lowest
amount practicable; to keep the levels that are discharged to
the river at a minimum; and to contain the high level wastes in
such fashion as to have definite and permanent control over them.
Handling Costs for Higher Level Wastes

The cost of storage including the tank farm piping and appurtenances is approximately \$1.60 per gallon. There are also smaller additional costs for evaporation, neutralization and handling of the waste at the separations facilities.

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#### DISPOSAL OF LOW LEVEL RADIOACTIVE WASTES

James M. Morgan, Jr. - The Johns Hopkins University
Survey of AEC Installations.

During the late summer and early fall of 1953, a group from the Department of Sanitary Engineering and Water Resources in The Johns Hopkins University made a survey of existing methods of disposing of low level radioactive wastes at some twenty AEC project installations in the United States. The purposes of the survey were: (a) to study methods of existing low level disposal practices; (b) to obtain actual cost figures of land disposal techniques and sea burial techniques. Since operations differ at each installation, it is not possible to compare either the costs or the methods of waste disposal, and no attempt was made to do so.

#### Findings of Survey.

All operations with radioactive materials, both in the laboratory and in production plants, produce a certain amount of low level radioactive trash or junk. This waste takes the form of combustible and non-combustible solids, i.e. "wipes", Kleenex, broken glassware and hardware and contaminated equipment. Liquids may also be expected.

The methods used for disposing of the low level wastes are:
burial on site, shipment to another site for burial, temporary
storage above ground, burial at sea, and incineration and confinement
of ashes. Geographical and/or environmental factors determine the
method selected in each case.

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The survey group found that eight installations currently dispose of waste by burial at sea. Four are on the Atlantic Coast and four on the Pacific. Commencing in New England and proceding clockwise arount the country, the installations employing sea disposal are:

Installation	Location	Disposal Site
Massachusetts Inst. of Tech.	Boston, Mass.	Massachusetts Bay or Atlantic Ocean
Brookhaven National Laboratory	Upton, Long Island	Atlantic Ocean off New Jersey
Westinghouse Atomic Power Div.	Pittsburgh, Pa.	Atlantic Ocean off New Jersey
National Institutes of Health	Bethesda, Md.	Atlantic Ocean off Vingania
North American Aviation, Inc.	Downey, Cal.	Pacific Ocean, Santa Gruz Basin
California Research and Devel.	Livermore, Cal.	Pacific Ocean, So. of Farallon Island
Univ. of California Radiation Lab.	Berkeley, Cal.	Pacific Ocean, So. of Farallon Island
U.S.Naval Rad. Defense Lab.	San Francisco, Cal.	Pacific Ocean, So. of Farallon Island

Sea Burial Practices: Most of the waste is disposed of in 500 to 1000 fathoms of water and no closer than 50 miles to shore. Since the inception of sea burial in 1946, approximately 8000 containers have been so disposed; about 2,000 packages in the Atlantic (including miscellaneous pails and small burial vaults) and 6,000 drums and concrete boxes in the Pacific. Drums used are either 30 or 55 gallon carbon steel containers. Through 1952, it is estimated that approximately 200 curies of liquid and solid waste have been dumped at sea.

Sea burial packaging is simple and effective. A very fluid concrete is poured around and over the dry wastes in open top, carbon steel drums. Liquid wastes and concrete are often mixed together

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roducing weeks however Louis Contraction In the drum. Sometimes liquid wastes in 5 gallon or smaller carboys are surrounded by concrete in the drums. When properly prepared, drums have a specific gravity of 1.3 to 1.5 to assure sinking.

Large, special packages take the form of reinforced, concrete boxes, rectangularly shaped, with capacities up to 80 cubic feet. One installation employs lead containers in the shape of cylinders to dispose of highly radioactive chips of metal.

Costs of Sea Disposal: For sea disposal, the Johns Hopkins
University survey found that when collection, handling, monitoring,
sorting, packaging, transportation, storage, loading, and unloading
were considered, the costs varied from about 20¢ per pound to slightly
above \$1.00 per pound for solid wastes. The lower figure fits
installations accumulating over 20 tons of waste per year, and the
higher value refers to smaller installations, those with less than 4
tons per year. The disposal of properly packaged, low level liquid
wastes at sea costs up to \$1.00 per gallon.

#### Summary.

Low level, radioactive trash accumulates daily at atomic industry installations. There is no reason to believe that the amounts will decrease. To protect and safeguard nearby residents as well as the plant workers, the release of wastes to the environment is carefully controlled. Current methods of burial at sea are expensive.

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OPEN DISCUSSION



#### GENERAL.

#### Health Aspects of Radioactive Waste Disposal

The problem of disposal of waste fission products differs

little from those met in large scale industrial waste disposal.

It has the peculiarity of not being susceptible to biological or (27)\* oxidative treatment. With a non radioactive toxic material, the toxicity increases as the concentration increases and mass action phenomena operate more effectively to remove the material. However, with radioactive wastes the toxicity is essentially unrelated to the (3) concentration of the element. The radioactive waste problem is like other industrial waste problems in one respect: production is not stopped because wastes are evolved. In industry, many alternatives must be considered, not only for the process as related to possible wastes, but also for the types of possible treatment for the wastes.

It should be kept in mind that the waste problem applies to domestic operations and to habitation generally. In practically every large city space for the cheap disposal of waste is strictly (27) limited, and becoming scarce.

Two Possible Methods of Disposal: It becomes necessary to seek out new types of treatment or disposal. For example, in the case of the Brookhaven National Laboratory, the radioactive waste output is not high (thousands of curies per year) but the disposal problem is most serious. These wastes cannot be discharged into the ground because the ground water is shallow and supplies the neighborhood.

The nearness of the Atlantic Ocean to BNL is a great advantage and (7) immediately suggests sea disposal.

\*Note- See key to speakers on page 79.

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In the future it may be that land disposal will look better than sea disposal, or vice versa, but it is premature to say now which is preferable. It may be that both will be favored; sea disposal for the ocastal installations and land disposal for mid(14)
continent plants.

# Work of the National Committee on Radiation Protection (NCRP) and Others.

The National Committee on Radiation Protection has studied the matter of ocean disposal of radioactive wastes and has prepared a handbook on the subject.\* However, the recommendations in this handbook refer to relatively small amounts of radioactivity. Definite recommendations, such as packaging wastes in concrete and dumping in depths 1000 fathoms or greater, were possible because of prior, (10) accumulated experience.

Definition of the Problem: One of the most prominent difficulties encountered in studying problems of this kind is that of semantics. Words such as pollution, containment and disposal have to be defined. Pollution: Pollution is the result of the accumulation of by-products of man's activity in the environment: land, air or water. The term applies when the accumulation has detrimental effects aesthetically or on human health. Pollution sometimes results in catastrophic changes in natural life in the area affected and it may be a cumulative event causing a gradual change over a period of up to hundreds of years. Containment: Containment is defined as storage in vats, pits or burial on land. Contained material is available for recovery or for relocation. As long as this material can be guarded from people, or vice versa, it is not a hazard to life or property. Disposal: In disposal, the waste material is released to the environment and is no longer available for recovery, relocating or guarding. Releasing to the ocean is an act of disposing; it is a final act; the material is not expected to (10)be recovered.

<sup>\*</sup> National Bureau of Standards, Handbook 58, "Radioactive Waste Disposal in the Ocean."

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and the second second by most and the cost of an area of the cost the section to the section of the se Hazards of Ocean Disposal: The NCRP committee resolved that there are 3 types of hazards to be considered in oceanic disposal of radioactive wastes: direct, indirect, and ecological. Direct hazards are those in which there is sufficient concentration of active material to involve danger to anyone coming close to the material, e. g. containers of radioactive wastes dumped at sea can be dangerous if they float. Indirect hazards are those involved in the reconcentration of radioactive wastes by organisms living in the sea, especially when these organisms ultimately may be used for human food. By biochemical concentration, the organisms in the food chain may reach high levels of radioactivity. An ecological hazard arises from radioactive materials being added to the sea. The radioactivity results in an unpredictable change of biological life in the ocean.

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## Radioisotope Concentration by Marine Organisms - Indirect Hazard:

Many of the fission product elements are present in natural sea water in trace quantities. However, the concentrations present are not known for more than a few. Strontium is one which has been studied in the natural environment. Most algae and other plants concentrate concentrate Sr by a factor of about 8. For those protozoa which are known to make skeletons of strontium sulfate, it is possible to estimate a concentration factor of about  $10^4$  on a wet volume basis. Generally, the Sr concentration factor from organism to organism ranges from 0 to  $10^4$ .

The weighted, mean concentration factor for the organism known to concentrate iodine is approximately 500,000, and in many instances this goes up to  $10^7$ . Nitrates are concentrated by dinoflagellates by a factor of about  $10^8$ . Each year organisms of biological importance are discovered to concentrate various elements which no one had previously suspected could be concentrated by such organisms.

Ecological Hazard: From the biologist's point of view, the pertinent factor is how high the radioactivity dumped in the sea can be allowed to go before it has physiological effects on organisms. Organisms will be affected, not by the radioactivity in the volume of sea water, but by the radioactivity in the volume of the organism. In the past few years the effects on large varieties of organisms exposed to continuous radiation over long periods have been studied. Some organisms are very insensitive and others very sensitive. The sensitivity extends to 10<sup>4</sup> on each side of the mean for a sample of

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great variety.

Eliminating Hazards of Ocean Disposal: Direct hazards
can be eliminated by dumping wastes in submerged packages in deep
water far from land where they would be practically impossible to
recover. These hazards may also be eliminated by diluting the
waste to a level that represents no hazard at all. Indirect and
ecological hazards can be avoided by keeping the radioactive materials
out of the biological and biochemical systems in the sea. These
hazards can be minimized by isolating and diluting the active
(10)
material.

Possible Tolerance Levels for Ocean Disposal: Allowable concentrations of radioisotopes in air and in drinking water were calculated by considering how much of water and air are consumed. By using a figure considered safe for a body burden it is possible to work back and calculate what the permissible concentration is for direct human contact. Going back a step further it is possible to figure how the marine organisms might eventually be used as (10) food by humans and thus represent a direct hazard.

At the present time it is not possible to even estimate what tolerance levels may be because there are too many unknown factors. Isotopic dilutions of elements in the sea, the amount of radiation a population can be exposed to without upsetting the population, the time of recycling of isotopes in the sea and the circulation of the sea which would determine volumetric dilution, are some of the unknowns which have to be determined before the safety of sea (3) disposal can be evaluated.

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## Need for Oceanographic Research.

If radioactive wastes are dumped in the ocean, sooner or later it will be necessary to face the public with some facts regarding the fate of the radioactivity. For this reason, even if the ocean has capacity for almost unlimited radioactivity for the next 100 years, when the wastes are dumped the whole area will have to be monitored.

At the present time, installations in the United States and Great Britain are dumping radioactive wastes in the sea. To be sure, these dumpings consist of "low level" wastes; however, little or nothing is (ed.) being done to ascertain the future state of the wastes.

Summary of Unknowns in Oceanography: Very little is known about the behavior of the ocean environment when foreign materials are introduced. Some of the radioactive materials may aid oceanographers in studying problems of ocean behavior; at least, they make it possible to learn a great deal beyond the immediate behavior of the radioactive material itself. Oceanographers would like to know more about the following problems in order to formulate recommendations concerning ocean disposal of radioactive materials.

- 1. The biochemistry of the elements in fission produce wastes:
  What are the effects on marine organisms? Will the elements affect
  the productivity of the organisms? Will the wastes affect the value
  of food which is taken from the sea?
- 2. The inorganic chemistry of the elements in fission product wastes: Do the wastes remain in solution or precipitate out? What is a desirable form for the wastes?

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3. The patterns and magnitudes of circulation of the oceans themselves. For deep and shallow waters it would be necessary to know the rates of movement, overturn and mixing to predict the fate (10) of any disposed material.

## Stability and Circulation of the Oceans.

The ocean is essentially a film of water on the surface of the earth; the vertical dimension is about one one-thousandth of the horizontal dimension. Most of the mixing that occurs in the ocean is not vertical but horizontal; this results from dimensional differences and from vertical density gradients. Convective overturn (10) is an exceptional phenomenon.

It was previously thought that the wind moved most of the ocean's waters. This appeared obvious. However, if there were no wind at all, the oceans would still circulate in much the same way as they do at present.

In each of the oceans there is a lens shaped layer of water with a relatively high vertical temperature gradient which lies deepest in mid latitudes, i.e. the surface layer is separated from deep water by this thermocline layer which happens to be thickest in mid latitudes and thinner near the equator. It outcrops at about 50° to 55° latitude in all the oceans; see Fig. A. In the southern hemisphere oceans there is apparently a good deal of sinking along surfaces of constant density into this "thermocline" layer because the water can be traced from the surface in high southern latitudes all the way to the equator. In the Atlantic the water

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can be traced across the equator; remnants of it are found at 1000 meters in the Gulf Stream.

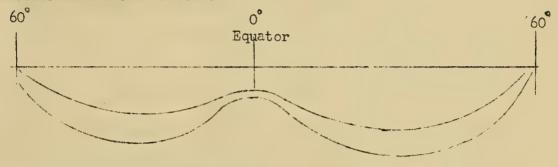


Diagram of Thermocline Layer

In the high southern latitudes another sort of water is produced which is much heavier than the aforementioned one. The heavy water sinks and goes to the bottom; it is the source of all of the bottom water. It dominates circulation in the Indian and Pacific Oceans. The pattern in the Atlantic is more complex. The Mediterranean Sea continuously dumps a considerable volume of very saline water into the Atlantic system. Mediterranean water spreads laterally away and can be traced into the South Atlantic at mid depths. In the high latitudes of the North Atlantic surface water is cooled, sinks and spreads south toward the equator, but it is not dense enough to displace the antarctic bottom water. However, it does stay below (16) the Mediterranean water.

## Circulation Rate in the North Atlantic.

The most evasive factor in determining the overturn rate is the amount of water that sinks in the northern latitudes and which moves toward the equator stirring things up rather gently. There are about 30 million cubic meters per minute being transported

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near the surface. (The Atlantic is essentially closed at the northern end.) Therefore, it must come back somewhere underneath. Actually, some of it travels southward quite near the surface, in the main thermocline, some of it a little deeper, but apparently only (16) occasionally does it come back via the bottom.

If the amount of water that flows into the North Atlantic across the equator at the surface (based on dynamic computations) is divided into the volume of the North Atlantic, the result is a time of circulation of the order of magnitude of 10<sup>3</sup> years, assuming that the circulation is perfect, that there are no eddys, dead (26) pockets or accelerated movements.

Institute of Oceanography of Columbia University has been working on a problem closely related to the circulation of bottom waters. This group has been interested in the distribution of sediments on the ocean floor and heat flow through these sediments. The C<sup>14</sup> dating method which is employed involves collecting a 100 gallon sample of bottom water and extracting the carbonate from this at sea. The carbonate sample is brought back to shore and a measurement is made of its radioactivity. Knowing its activity establishes the age of the C<sup>14</sup> and presumably the age of the bottom water from which it was taken. Preliminary determinations by this method indicate the age of the bottom water to be of the order of magnitude of 10<sup>3</sup> years.

The Lamont Institute also has been making tritium assays of ocean water. It is anticipated that these H<sup>3</sup> measurements will indicate how much the annual mixing penetrates to below the seasonal thermocline.

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Tracer Technique Interferences: The disposal of fission product wastes in the sea would constitute no direct threat to the C<sup>14</sup> method. C<sup>14</sup> could still be measured even after a lot of contamina-(9) tion has been put in, but no so with tritium. The interferences of C<sup>14</sup> particles which fall from the atmosphere, get into the sediment and mix in the bottom water depends upon how much the C<sup>14</sup> content of (26) the water is changed.

Oxygen Concentration Method to Determine Circulation: There are some oceanographers who do not like to believe that it takes thousands of years for the vertical overturn in the North Atlantic. There is just too much water being transported to high latitudes and too little of it coming back from high latitudes at the surface to permit such low rates of exchange. Formerly it was thought that almost every winter some surface water was made heavy enough to go to the bottom, but now it is realized that it does not have to go all the way to the bottom. If just part of the system is overturned, the overturn may actually be quite rapid. A different approach to the vertical overturn rate was discovered recently by Valentine Worthington of the Woods Hole Institution.

Worthington noticed that all of the deep ocean oxygen concentration measurements made by North Atlantic explorations 30 years ago showed more oxygen in the deep water than the recent measurements.

Both times there were a considerable number of stations covered and measurements were made very carefully. In comparing the two sets of data it has been found that they both agree in the shallow waters; however, below 2500 meters there is about 0.3 cc. of 0, per liter less

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than there was 30 years ago, apparently over the whole Atlantic Basin. This discrepancy indicates that oxygen is being used up at the rate of about 0.01 cc/liter/year. Using this rate to calculate the age of the Atlantic water below 2500 meters, a time of about 140 years is determined. The chronological weather record shows that a very great cold spell was experienced over the whole northern hemisphere 140 years ago.

Water has to be chilled to a lower temperature (below 3° C.) than the bottom water in order to displace it. In normal winters. 30 water is produced at the surface. It sinks but stays above the colder bottom water. One can imagine that a series of very cold years will produce dense water that will sink to the bottom. Apparently 1810 to 1814 was such a period; since then there has been no water made that has been heavy enough to get below 2500 meters. According to this theory the water below 2500 meters has been locked off for 140 years. It may very well be that the natural state of the ocean is to have no oxygen in the deep water.

Discrepancies between C14 and Oxygen Turnover Rates: One explanation why the C14 results to date have shown such large values may be that the C14 samples have all been taken very close to the bottom. From acoustical evidence it is pretty certain that something unusual happens near the bottom. It is believed that there is a turbulent mixing layer adjacent to the bottom. This layer would have available all of the carbon that the worms have brought up out of the mud. Critical age determinations by the C14 method should be made in the span between 500 meters above the bottom up to the bottom of the main thermocline.

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[4] C. Company, Sp. Phys. Lett. 19, 200 (1997).
[5] S. Company, S. Company, S. Company, Phys. Rev. Lett. 19, 120 (1997).

On the other hand, the diminishing oxygen concentration theory is not without possible fault either. Now that the earth as a whole is better covered with weather stations and climatic "cycles" are becoming better known, it is doubtful if in recent centuries the northern hemisphere could everywhere be below normal in temperature. Actually, cold areas are usually separated by areas of normal or above normal temperatures. The famous "year without a summer," 1816, observed in New England was part of the cold decade, 1811-20, which affected not only New England but Western Europe, including England. I have seen no data to indicate if the North Atlantic was also (37) cold.

Biological Interpretation of O<sub>2</sub> and C<sup>14</sup> Turnover Rates:

Marine animals are distributed at all depths in the ocean. Since bottom animals must have a source of organic matter to survive and the only known source of organic matter is in the surface layers, there must be a continuous filtration of organic matter down to the bottom. Biologists have not been able to measure this change in terms of the loss of essential nutrients from the surface layers.

If the rate of oxygen consumption in the deep water is about 0.01 cc per liter per year, it would take about 500 years to exhaust the oxygen. If this oxygen consumption rate is correct, it means that the rate of metabolism in the deep water is 1/500ths of the rate of metabolism in the surface water. Analytical chemical methods are not developed sufficiently to measure the biologically active materials to the order of magnitude required to bear this out.

However, if the organic matter is being formed at the surface, it too is contributing to the carbon cycle. The oxygen consumption

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rate indicates the rate at which the carbon is being contributed to the deep water -- a possible consideration when interpreting the  ${\rm C}^{14}$  (18) dating results.

Minimal Turnover Rate from Phosphorus Cycle: The quantity of phosphorus in the oceans is known within better than 50 percent. There are estimates of the productivity of the surface layers of the ocean, given in terms of carbon, but which can be converted into units of phosphorus. If all the phosphorus went through the life cycle, and was incorporated into plants when it came from the deep water, it would take about 500 years for all of the phosphorus in the oceans to pass through the productive layers once. This estimate gives an idea of the minimal turnover time because there is a great (26) deal of recycling of phosphorus near the surface.

#### BULK DISPOSAL

From the financial point of view bulk disposal of radioactive wastes looks more promising than does disposal of packaged wastes.

The oceans do have a large volume of water available for dilution. It remains for the physical oceanographers to tell how bulk disposal can be accomplished and how soon the radioactive wastes become mixed (27) and diluted.

# Ecological Cycle as Limiting Factor.

It seems that the ecological cycle is the limiting factor in sea disposal. It is not the dilutions possible but the biochemical concentrations which will limit bulk disposal, i.e. one organism will concentrate to a factor of 10<sup>4</sup> and a different one will consume the first organism and produce a further concentration of 104 for a total, say 108 in two jumps. This may happen with a few elements but not with many of the fission products. Most of the isotopes of concern will not be reconcentrated most will be rejected when the second organism eats the first one. However, the limitation imposed by the ecological system does not mean that a status quo situation must be maintained in the sea. There may be very marked changes in the ecological system without significant losses to the world. The present system represents equilibrium for the present set of conditions; if the conditions are shifted a little, the system adapts itself naturally.

#### Model Experiment Feasibility.

It may be possible to run a model experiment to study ecology.

It is almost impossible to duplicate the ecology of the sea but it

can be approximated enough so that a volume of water could be treated

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with a given amount of mixed fission products to determine what the general shift in ecology will be. There are difficulties with mixed fission products, however, for as soon as the volume in which they are placed is delimited they tend to accumulate on the walls. Most of the isotopes of importance in disposal are present in sea water as radiocolloids; these are strongly adsorbed by any available surface. In the sea the available surface is largely organisms, but in a defined laboratory system the available surface is mostly the enclosing walls. The next possibility is to use a small sized ocean like the Caribbean or the Mediterranean, for it would not be feasible to mark out for study some volume in the open ocean. Practical Experience with Acid Iron Wastes.

A barge load of liquid acid-iron wastes was discharged in surface waters under experimental conditions to determine whether or not the wastes had a detrimental effect on the waters. The total quantity of waste, 3000 tons of a solution of ferrous sulfate in sulfuric acid, was discharged in a single operation. The waste had about 300 tons of sulfuric acid and about 300 tons of ferrous sulfate in the one load; the density was 1.25. A dilution of one part waste to 1000 parts water makes it the same density as sea water. This dilution of 1:1000 occurred within a few hundred yards of the disposal barge. It was not possible to distinguish the difference between the turbulence due to the passing of the barge and the normal surface water turbulence. With the accuracy of the iron analysis employed, it was possible to trace the wastes to a dilution of 108. At one time a spotting plane was used and the men were able to trace the wastes

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for 12 hours after which they just disappeared below the 10<sup>8</sup> dilution. The acid wastes could never be traced deeper than 100 feet in water of 1.021 density.

From this experience it appears improbable that bulk liquid wastes discharged at the surface will go deep. Even with densities of the order of magnitude of those of liquids, the mixing would be rapid enough to keep the waste near the depth of discharge. If wastes are discharged at the surface it is to be expected that they will stay in the photic zone and be subjected to all the biochemical reactions,

To sum up the barging operation, the following phenomena can be expected to occur:

- a. Immediate mixing behind the barge.
- b. Development of waste concentrations in the diluting medium following the immediate mixing stage.
- c. A long term accumulation which would be of primary concern in bulk disposal of radioactive wastes. (26)

#### Disposal as an Experiment.

The surface waters may be the proper place to dispose of increasing amounts of waste. A real answer to whether or not bulk disposal can be accomplished is to try it at a safe and low level to get enough information to be sure of what is happenning. If limited amounts of liquid waste were released from a tanker or barge to surface waters, the wastes might enter the biological chain rapidly and could possibly be traced. If it is found that the activity levels were creeping up to 2% or 3% of what was permissible, then the dumping practice could be stopped or reduced to correct the situation. This type of bulk disposal amounts to a super tracer experiment. At (7) the same time it disposes of a quantity of wastes.

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Effect of Ocean Turnover on Waste Disposal: According to
the dissolved oxygen theory of turnover, if radioactive wastes are
dumped in the high latitudes in the winter time, they would not go
to the bottom but would sink to around 2500 meters because the water
produced annually in the high latitudes goes to that depth. If the
material is put below 2500 meters it might stay there for a very long
time or at least until there are winters colder than those of 1810
(16)
to 1814.

#### Possibility of Sargasso Sea for Bulk Disposal.

Perhaps the best place to discharge radioactive wastes in bulk would be into the Sargasso Sea. It contains very few marine animals and plants. Based on the barging experience with the acidiron wastes, the contamination should be localized. The most damaging concentrations would be behind the barge for a limited distance, which is small compared to the whole sea. The waste material will go into a lens of water at the surface which follows the circulation pattern of the Sargasso. A drift bottle takes 1 to 3 years to circle around; it is expected that the waste will drift around in about the same length of time. Some of the waste material will be sloughed off at the edges into areas of mixing. This will be of concern only from the point of view of long term accumulations in the ocean; here again it is desirable to know the pattern and time of circulation for the whole ocean.

To sum up, for bulk disposal of radioactive wastes into the Sargasso Sea it is necessary to consider: (a) the annual rate of pollution, (b) the rate of dispersal of the wastes in the Sargasso

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Sea and of the Sea into the Atlantic Ocean and (c) long term (26) accumulation and its effects.

# Radioactive Tracer Experiment Possibilities.

A radioactive tracer experiment should furnish sufficient information about the diluting capacity of the ocean and about the (6) effect on marine organisms. The first thing to determine is what can be learned in any experiment. In a deep water experiment you can learn about rates of horizontal and vertical diffusion and perhaps something about currents, either tidal or net drifts. In an experiment nearer the surface the same things can be learned, in addition to something about the rate of transport through the main thermocline which ultimately will be important and necessary to sample. Many experiments will be required before any definite conclusions are (18) reached.

A proper experiment may be one wherein a large quantity of radioactive material is placed in bottom water. The movement there should be slow enough to enable tracking for a long time unless some (9) organisms precipitate out the activity.

Magnitude of Experiment Required: Navigation or precise certainty of position determines the size of the tracer experiment required. The contaminated area cannot be smaller than 2 miles on a side or it will not be found. There should be enough radioactivity to cover an area 50 miles in diameter at easily detectible levels at the time results are measured. A 50 mile area can be navigated (35) to and is small enough so that it will not get out of hand. One ship can make a survey of this sort. It will be necessary to set up

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(16)

an auxiliary navigation system to determine the exact location.

Anchor buoys will have to be spotted around to navigate from,
especially during the early stages when the contaminated area is
small and can be followed around with detectors. When the activity
spreads out it will not be difficult to spot, but the activity readings will be close to the limits of detection. It will not be easy
to determine relative concentrations and concentration areas at this
(25)
time.

Radiation Measurement Difficulties in Bottom Waters: The practical design limit for a tracer experiment in the deep open ocean appears to be the ultimate sensitivity of the radiation measuring The readable level is about 10<sup>-19</sup> megacuries per cc. Instantaneous reading of radioactivity may be obtained by sending down a sensing instrument. The limiting level for this technique is about 10 megacurie per cc. If a volume 100 kilometers (62.5 miles) square by 100 meters thick received one megacurie, the activity would be diluted to  $10^{-7} \times 10^{-7} \times 10^{-4} = 10^{-18}$  megacuries per cc. -- well below the direct or instantaneous readable level. Taking samples for concentration and subsequent reading introduces many difficulties. In such a case you would not know where to take the sample, nor would you know whether you had selected the right location for sampling until sometime later. If a smaller area was charged, direct readings might be feasible. The material would start diffusing the instant it was released and tracing it with a direct reading instrument should give information as to how fast diffusion takes place at the bottom. Nobody knows how fast it would be, although observations are available for

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surface conditions. Rapid diffusion should be an asset in diluting (18) the radioactivity.

Energy Absorption and Upwelling: When a megacurie is put in the ocean it may heat up the water and possibly cause upwelling. The heat dissipation will probably contribute to the waste's dilution by (5) (14)(32) making the water move. Some comment that this is no problem, that the heat release rate is low and that the heat will dissipate (ed) quickly.

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The dilution caused by the heat emission of the wastes depends a great deal upon the density gradient of the ocean at the site.

The density gradient near the ocean bottom is not accurately known; oceanographers have been trying to measure it for years. The difficulty lies in the fact that thermometers cannot be placed at precise depths (16) measured from the bottom.

Sampling problems: It is very difficult to sample at short distances above the bottom. The technique of handling instruments at precise distances above the bottom is not yet sufficiently well developed (16) to proceed with bottom layer measurements, although the Lamont group has worked out a system whereby samples can be taken near the bottom in conjunction with their core sampling device. A mechanism on the corer trips the water sampler or electrical thermometer attached to the device. By this procedure the water is sampled at 30 feet from the bottom. It would be possible to approach closer to the bottom (9) but it would be more difficult to get a core at the same time.

Radioactivity readings may be taken instantaneously by sending down a sensing instrument. This procedure should be feasible if the concentration is high enough. The limiting level is about 10<sup>-7</sup> microcuries per cc. However, if the instrument is used for continuous underwater measurements it will absorb radioactivity and this will result in continued readings even after the activity has dispersed.

This trouble can be avoided by leaving the instrument down only long (14) enough for the measurement.

Adaptable Instrument: There are instruments similar to those used for mine hole logging under water pressure which might be extended

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to this operation. (The radioactivity levels at which the gamma, well logging instruments operate is many orders of magnitude higher than natural radioactivity in the ocean. Generally in the well work, they irradiate the side wall of the well with a particular radiation and then look for a certain reaction. They do not have to work at (9) low levels. ) The instrument is pressure packed with grease to make it rather solid on the inside; it can now operate under about 2000 ft. of water. It is connected by a shielded, mono-conductor having a woven stainless steel jacket to an Esterline Angus Recorder which makes the records. Power is sent to the instrument circuit from the (14) surface and the signal comes back up the same conductor.

Probing Problem: The depth of the well logging instrument is measured by length of cable out on a spool. In the sea application this method would tell only how much cable is out and not where the (14) instrument is situated, i.e with 20,000 feet of cable out the location of the instrument in the lowest 200 to 300 ft. would be (32) uncertain. The waste concentration is expected to vary with depth (16) as well as spread out laterally.

An echo sounder device on the detector may help pinpoint the location. The Woods Hole Institution has operated echo sounders at 2000 feet depths and are continually striving for greater depths.

However, if an echo sounder is put on in an attempt to work from the bottom, there would still be a difficult cable problem because the echo sounder has to be driven. All the instruments together must also have a very small physical bulk; otherwise the cable will drift (16) laterally to one side of the ship.

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Cable Problems: The cable for the well logging instrument may be strong enough for a depth of 2000 feet in a well but it may not be adaptable to 20,000 feet of depth in the sea. The Woods Hole group has not been able to persuade a cable manufacturer to build a cable with an electrical lead in it that can be guaranteed to be safe to lower more than 20 or 30 times. In oceanographic work the cable goes down under terrific tensions and when it comes back up the lead tends to creep inside the cable. Eventually the cable breaks the lead. The cable used in the oil industry (well logging) just will not work at sea because the ship heaves on it; the winch moves up and down quite rapidly as the ship rolls and pitches in the seas. The strains are much higher than those faced in the oil industry.

By-Passing Cable-Conductor Problem: If it is necessary to have the electric circuit at the bottom it may be possible to send the power down with the instruments. Then only a regular cable would be needed to lower and raise the measuring devices. The inclusion of a recording depth element would provide the essential depth records. Similar instruments have been used to measure radioactive elements in the atmosphere. The instruments are sent up in baloons and telemeter their information back to the surface; perhaps the same set up can be used either with a telemeter circuit or with a recorder circuit.

Mid-Depth Experiments: Many of the deep water experiment

problems may be eliminated by working in shallower water, i.e. in or

(5)

below the thermocline. The evidence shows the quietest waters

are at mid depths, not at the bottom. At mid depths the experiment

is likely to run for the longest time. To get the radioactive material

(16)

to these depths may be a problem. However, mixing may be rapid

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(25)enough so that waste density would not be significant. The sampling problem may become more difficult if the material diffuses downward whereas on the bottom one dimension would be delimited. The sampling problem may be alleviated if the radioactivity can be measured directly. possibly by towing the instrument through the area of activity much like the mid water trawl is towed. Depending on the speed of towing a length of cable as much as 3 times the depth Wertine of has a photomultiplier might be needed. which can go to a 6000 ft. depth. If a scintillation cell is put in front, it would be readily adaptable to this experiment. Plate power is sent down from the surface. Again, however, if the instrument is submerged continuously the walls of the cell are likely to pick up radioactivity but it may be that stainless steel window and walls (to measure gammas) will alleviate that difficulty.

Convection in Surface Waters: In the ocean surface water is being cooled more than half the time in the winter months. In the summer months it is being cooled less than half the time by evaporation in many areas. This cooling sets up a complicated pattern of motion (convection cells) moving the water along in a helical pattern. When well developed, the convection cells are very large and vary in size with changes in wind velocity. Any surface experiment will require (16) much study to account for all of the large scale motions that exist.

Place and Time for Experiments: In the Sargasso Sea area hurricanes are a source of worry particularly in the autumn. In the spring the surface layers are rather stable over all of the Sargasso Sea except on days when the winds blow full force. It may be better

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to go down to the trade wind regions where there is a steady wind and more or less constant conditions throughout the year.

In the winter in open ocean the water is actively overturning down to about 200 meters. In the summer on a typical day it overturns down to about 25 feet, almost a whole order of magnitude (16) difference.

### Instrumentation for Backgrounds.

Instrumentation is a problem in any of those ocean experiments.

Instead of thinking of instruments for a tracer experiment, it may clarify things to think of instruments necessary to determine the natural radioactivity in the sea. If instruments were developed for (10) backgrounds, then they would be available for the other purposes.

If instruments were available they could be put aboard any ship
going out and could take background measurements as part of the other
(14)
work. Among all the oceanographic laboratories there are ships
going generally in all directions. If 4 or 5 instruments were available,
it would be possible to essentially cover all of the world's ocean
(16)
areas and all types of water.

Work of Lamont Institute: To establish natural backgrounds for several types of radiations as a function of depth in the sea, the Lamont Institute group decided it would be easiest to utilize photographic plates loaded several different ways. The plates are fine to establish natural backgrounds, but they are not worth much as an instrument in the diffusion experiments because they would not give the information fast enough.

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### Fluorocein Dye Experiment Possibilities.

A dye, like fluorocein, may be easier to follow than radioactivity in tracing diffusion and current effects in an oceanic
(9)
experiment. A dye experiment should be done independently of
other experiments because it will answer questions such as those
regarding small scale turbulence. If the turbulence spectrum in the
(25)
sea was known, it would facilitate solving other problems. The
use of dye in an experiment preceding the radioactive tracer experiment appears to be a good idea because it would be difficult to handle
(6)
a megacurie of activity the first time. If dye was used it could
be used as a dry run for the radioactive experiment, i.e. the dye
could be handled with the same equipment that would be used for
(5)
handling the megacurie of activity.

Experience with Fluorocein Dye: Several pounds of fluorocein released at depths of 50 feet or more did not become visible at the (35) surface of the ocean. In another experiment in an estuary of the Chesapeake Bay 50 gallons of concentrated dye was pumped in  $2\frac{1}{2}$  minutes into water 17 feet deep. In 15 minutes the dye had been diluted by a factor of  $10^5$  and in 60 minutes by a factor of  $10^6$ . It was not possible to trace it further with the equipment available. It appears possible to be able to trace very large dilutions of fluorocein. However, there may not be enough fluorocein made in this country to run an experiment in the open ocean in a volume 10 miles (25) square by 100 feet thick. From previous experiences, working with relatively small amounts of fluorocein, the isothermal layer was thick enough to permit expansion in 3 dimensions. It may take

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something like a few tons to fill up the fairly shallow isothermal layer to get a sufficient area covered (for ease of navigation).

With ton quantities of dye perhaps the initial three dimensional (35) expansion approaches two dimensions.

Equipment for Dye Experiment: Since natural sea water is

fluorescent there is a background problem to solve in any fluorocein
(25)

dye experiment. There is little information on the spectrum of
(5)

natural fluorescence and the spectrum of fluorocein fluorescence.

Equipment for directly measuring dye tracers is non-existant and would
(25)

have to be built.

A dye measuring instrument is conceived to be a photo multiplier
(5)

type of device with an ultraviolet light source. The dilute
fluorocein must be brought into essential contact with the tube

possibly with a window that would pass the visible light and not the

ultraviolet. Existing laboratory instruments may be adaptable to
(25)

the field work.

There may be substances other than fluorocein dye which have better ultraviolet fluoresing properties. In normal photo comparator techniques it is better to use an ultraviolet light source for fewer (5) compensations have to be made. The best material would be that which fluoresces in a wave length not reproduced in a natural (25) environment.

#### Other Diffusion Experiment Possibilities.

The Woods Hole group is developing a device which can be put down in the deep ocean for very long periods of time. The essential feature is a release device which can be activated from the surface

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ship. The instrument can be floated above a mooring at mid-depths to measure movements there. When it is desired to pick it up, an explosion signal floats it up with its records. This device is working successfully. The collection of data will start shortly.

Diffusion Experiment with Drift Bottles: One great weakness of the radioactive and dye tracers is that they become too diluted to measure. Drift bottles do not. For intermediate depth experiments bottles or other types of drift particles have to be placed at the desired depth to remain there until measured some time later. It may be possible to develop an oceanographic version of the constant altitude balloon used atmospheric work. In order to keep track of the subsurface floats, time bombs on them could be fired and the (32) positions determined by a SOFAR network. Drift bottles would not give the small end of the turbulence spectrum as well as a tracer (25) but they should at least give the large scale end.

Dr. Parr of the New York Museum of Natural History suggested that the distribution of the Caribbean might be followed with some of the foraminifora. These organisms grow in the tropical sea and are transported out in the currents. Perhaps Carribean plankton (27) or some similar indigenous material be used.

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#### DISPOSAL IN SEDIMENTS

### Containing Packaged Wastes in the Sea Bottom.

There exists the possibility of using sediments on the sea floor as an enveloping medium. If engineering progresses so that waste volumes may be economically concentrated, the sea bottom becomes a logical place for disposal. On the bottom the wastes may in time be covered with sediments with reduced possibility of dispersal.

A suitable container of reasonable size to carry the concentrated wastes down might be 2 feet in diameter by 20 to 30 feet long with a mean density of 3 or 4. The container should sink through the water with a velocity of at least 60 knots; with a carefully selected bottom it might penetrate one length. A proper location would be in a basin having active deposition which would cover the container.

In reviewing the idea of disposal in the bottom with geologists it was learned that, generally, they are not too sanguine on the penetration theory but were quite hopeful on the possibility of areas with rapid deposition. Unfortunately none of those areas of deposition are close to existing waste producing plants. Transportation (27) and cost of transportation are matters of no small concern.

### Natural Adsorption on Sediments.

There are natural processes occurring at river mouths which remove from water many of the elements in the fission product range.

If this adsorption did not occur, many of the elements present in sea water in trace quantities would be present in much larger amounts.

Because of the possible deleterious effects on sea life along the

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coast line, it is not feasible to discharge radioactive wastes at river mouths. However, it may be possible to take advantage of sediment adsorption at sea; to lock the fission products in similar sediments and deposit them on the ocean floor, again in a basin with (35) active sedimentation.

The pH of rivers is conducive to uptake by silts. However, if the silt comes to an estuarian system that moves into the ocean, (25) the pH change would effect some desorption. In the normal ion exchange mechanism it is likely that sodium will displace, to some (8) extent, the material adsorbed on the clays.

Experiments with Silts and Sediments: A Johns Hopkins University group studied the adsorption of Cu, Sr, sulfate sulphur, P, Fe<sup>†††</sup> and iodide ion on silts and sediments from various natural sources and also on some commercial products. With all of these ions, except I, a pH range and a total salt concentration range were found for which these materials were rapidly and rather completely adsorbed. The iodide ion was not taken up appreciably by any of the adsorbers.

Approximately 40% of the adsorbed P was lost in going from a fresh (5)
water stream situation to a marine situation.

Practical Experience at Oak Ridge: Since the start of operations, the Oak Ridge National Laboratory has been discharging low level radioactive liquids into White Oak Creek. The Creek runs into a retention basin before being drained into the artificial White Oak Lake and later into the Clinch and Tennessee Rivers. Water in the controlled retention basin is checked for activity level before release. Most of the radioactivity dumped in the basin is deposited in the clay

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of the basin itself. In monitoring the Clinch River System,

most of the radioactivity has been found in the bottom and in places
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where there is deposition. The radioactivity on the bottom

increases behind dams where there is deposition and is practically
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nonexistant where there is scouring.

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### Summary.

From the discussions at Woods Hole on August 6, 1954 it was clear that recommendations regarding sea disposal of radioactive wastes could not be made. The uncertainty regarding safety of sea disposal stems from the fact that the physical, chemical and biochemical behavior of the oceans cannot be satisfactorily predicted. Rates of circulation, diffusivity and turnover in the oceans have not been firmly established.

There are many technical aspects to be explored even before experiments can be designed to evaluate the possibilities of sea disposal. The greatest drawbacks appear to be: (a) the lack of suitable electric conductor-cables to go deep in the ocean and (b) the lack of suitable instruments to measure low levels of activity under water.

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### Key to Speakers

### Conference on Disposal of Radioactive Waste

Woods Hole Oceanographic Institution Woods Hole, Massachusetts

August 5-6, 1954

	Name	Position	Representing
1.	Arnold Arons	Assoc. in Physical Oceanography	W.H.O.I.
2.	Hanson Blatz	Chief, Radiation Branch	A.E.C., N.Y.O.
	V. Bowen	Geochemist	W.H.O.I.
4.	Dean F. Bumpus	Oceanographer	W.H.O.I.
	Dayton E. Carritt	Asst. Prof. of Oceanography	C.B.I, J.H.U.
	Walter D. Claus	Chief, Biophysics	A.E.C., Wash.
7.	F. P. Cowan	Head, Health Physics Div.	B.N.L.
8.	F. L. Culler	Dir., Chemical Technology Div.	O.R.N.L.
9.	Maurice Ewing	Geophysicist	Columbia Uni.
10.	R. H. Fleming	Exec. Off., Dept. of Oceanog.	Univ. of Wash.
	F. C. Fuglister	Oceanographer	W.H.O.I.
	J. C. Geyer	Prof. of Sanitary Engineering	J.H.U.
13.	A. E. Gorman	Sanitary Engineer	A.E.C., Wash.
14.	W. B. Harris	Industrial Hygiane Engineer	A.E.C.
15.	L. P. Hatch	Sanitary Engr., Nuclear Engr. Dept.	B.N.L.
16.	C. O'D. Iselin	Senior Physical Oceanographer	W.H.O.I.
17.	A. B. Joseph	Research Staff Assistant	J.H.U.
18.	B.H. Ketchum	Senior Biologist	W.H.O.I.
19.	J. A. Lieberman	Sanitary Engineer	A.E.C., Wash.
20.	E. W. Moore	Prof. of Sanitary Chemistry	Harvard
21.	J. M. Morgan, Jr.	Research Staff Assistant	J.H.U.
22.	D. R. Norton	Chemist	W.H.O.I., U.S.G.S.
23.	Orlando Park	Consultant	O.R.N.L.
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26.	A. C. Redfield	Assoc. Director	W.H.O.I.
	C. E. Renn	Prof. of Sanitary Engineering	J.H.U.
	F. A. Richards	Chemical Oceanographer	W.H.O.I.
	J. H. Ryther	Marine Biologist	W.H.O.I.
	E. H. Smith	Director	W.H.O.I.
	Paul F. Smith	Oceanographer	U. of Miami, WHOI
	H. Stommel	Oceanographer	W.H.O.I.
	E. G. Struxness	Sec. Leader, Health Physics Div.	O.R.N.L.
	T. T. Sugihara	Chemist and Radiologist	W.H.O.I., Clark
	Allyn C. Vine	Oceanographer	W.H.O.I.
	L. P. Wagner	Oceanographer	U. of Miami, WHOT
	Harry Wexler	Meteorologist	U.S.W.B., Wash.
	Edwin Wiggin	Mgr. of Tech. Information	Atomic Indus.Forum
39.	Joe Clark	Sanitary Engineer	S.R.P., duPont

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